



Impact des variations climatiques sur les ressources hydrogéologiques

Antoine Armandine Les Landes

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pour le grade de

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présentée par

Antoine Armandine Les Landes

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Géosciences Rennes

UFR Sciences et Propriétés de la Matière

**Impact des
variations
climatiques sur
les ressources
hydrogéologiques**

**Thèse soutenue à Rennes
le 21/11/2014**

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A mes grands-parents et parents,

Résumé

Les eaux souterraines représentent environ 98% des eaux douces potentiellement disponibles pour l'homme sur notre planète, en faisant donc une ressource fondamentale de nos sociétés. Du fait du changement climatique et du fort lien existant entre le climat et le cycle de l'eau et donc les ressources en eau, l'identification de la sensibilité des eaux souterraines aux variations climatiques est devenue primordiale. Or, le climat est un système hautement complexe dont les variations, contrôlées par de nombreux facteurs à la fois naturels et anthropiques, s'effectuent sur toutes les échelles de temps. Les modifications climatiques et leurs impacts sur le cycle hydrologique ne sont donc pas un phénomène nouveau. Les eaux souterraines ont par conséquent subi dans le passé les effets de ces variations climatiques, elles en subissent actuellement les changements et subiront celles à venir. Les travaux développés dans cette thèse ont abordé cette problématique visant à caractériser la sensibilité de la ressource en souterraine face aux variations climatiques. À l'aide de l'analyse de traceurs environnementaux à l'échelle des aquifères du Massif Armoricaïn, les impacts d'évènements climatiques majeurs intervenus sur les derniers millions d'années (transgression marine et période glaciaire) sont mis en évidence au sein du système hydrogéologique actuel. Ces informations paléohydrogéologiques permettent d'apporter de nombreuses contraintes sur l'hydrodynamique des aquifères du socle Breton sur une grande échelle de temps et constituent notamment une solide base pour des études numériques. Puis, à l'aide de la modélisation numérique des écoulements à densité variable, les grandes étapes paléohydrogéologiques liées à la salinité ont été investiguées, à travers les mécanismes de salinisation des aquifères par les rivières et le lessivage de l'empreinte marine. Enfin, dans un contexte un peu différent (échelle spatiale et temporelle), une modélisation numérique hydrogéologique a été édifée afin d'étudier la réponse des zones humides alimentées par un aquifère sous-jacent face à différents scénarios d'évolution du climat régional et à l'utilisation de la ressource en eau souterraine. Toutes ces études mettent en avant la haute sensibilité des ressources hydrogéologiques aux variations climatiques et hydrologiques, en termes de qualité et de quantité. La gestion de cette ressource nécessite donc de considérer à la fois les risques climatiques et non climatiques ainsi que de prendre en compte l'adaptation à long terme de ces systèmes.

Mots clés : Hydrogéologie, ressources en eau, climat, impact, modélisation numérique, traceurs environnementaux, zones humides, rivières, océan, salinisation.

Abstract

Groundwater resources represent approximately 98% of global freshwater resources available for humans on our planet; therefore groundwater is a fundamental resource for our societies. Due to climate change and the strong link between climate and water cycle and so groundwater resources, an understandable concern has appeared about the potential impacts of climate change on water resources. The assessment of groundwater sensibility to climate variations has become essential. Climate is a highly complex system with variations driven by many factors, both natural and anthropogenic, occurring on all time scales. Climatic changes and its impacts are not a new phenomenon. Groundwater resources have already been impacted by effects of climatic variations, being impacted presently and will be in the future. The work carried out under this thesis covered this issue to characterize the groundwater resources sensibility to climate variations. With the help of environmental tracers available at the regional scale, impacts of major climate events that have occurred during the last millions of years (marine transgression and glacial period) have been identified in the current hydrogeologic system. These paleohydrologic informations provide constraints on the hydrodynamics of Brittany aquifers on large time scales. Afterward, supported by density-dependent groundwater flow modeling, the major paleohydrogeologic steps related to the salinity have been investigated through mechanisms of groundwater salinization below coastal rivers and the subsequent leaching of marine waters. Then, in a slightly different context (spatial and temporal scales) a hydrogeological modeling was edified to study the impact of climate change on wetland extension due to the specific relationships between groundwater and surface water bodies. All these studies highlight the important sensibility of groundwater resources to climate and hydrologic variations in terms of quality (salinization) and quantity (lower water levels). Thus groundwater resources management needs to consider both climatic and non-climatic risks and the long-term adaptation of hydrogeological systems.

Keywords: Hydrogeology, groundwater resources, climate, impact, numerical modeling, environmental tracers, wetlands, rivers, sea, salinization.

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Chapitre 1. Introduction

1.1. Climat et eaux souterraines

L'eau souterraine constitue une ressource naturelle, fondamentale de nos sociétés (Hiscock, 2005) et viable pour de nombreux secteurs d'activités humaines: agriculture (42%), domestique (36%) et industrie (27%) (Döll et al., 2012). De plus, elle représente presque la moitié des eaux potables dans le monde (UNESCO, 2009) et joue un rôle très important dans le fonctionnement des rivières, des lacs, des zones humides et des écosystèmes (Treidel et al., 2011). Ses caractéristiques propres, telle que sa nature souterraine et donc « non visible » en font une ressource difficile à gérer contribuant ainsi à l'augmentation des pressions sur cette ressource. Or cette ressource en eau souterraine fait partie intégrante du cycle de l'eau. Le cycle hydrologique (Figure 1) décrit le mouvement continu de l'eau entre les différents réservoirs d'eau présents sur Terre : l'atmosphère, les rivières, les zones humides, les glaciers, les océans, les sols et les roches. Les transferts d'eau entre ces différents compartiments ont comme moteur l'énergie solaire. Sous l'effet de cette énergie, l'eau change continuellement d'état, entre liquide, solide et gaz, permettant ainsi les échanges d'eau entre les différents réservoirs. Il existe donc un fort lien entre le climat et le cycle de l'eau.

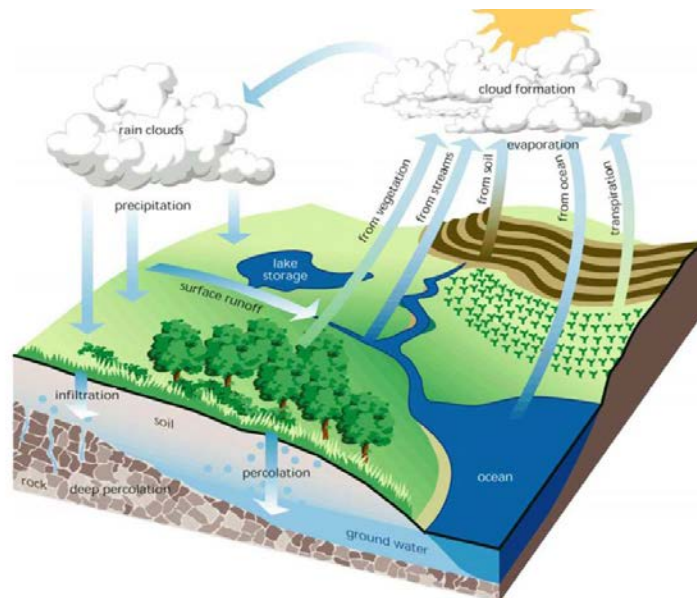


Figure 1. Cycle de l'eau (<http://www.nrcs.usda.gov>)

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Le climat est communément défini comme étant une météorologie moyenne et plus rigoureusement comme une description statistique (moyenne, variabilité, fréquence, amplitude, tendances... (Cubash et al., 2013)) en termes de paramètres pertinents (température, précipitation...) sur des périodes de temps s'étalant du mois aux millions d'années. Le climat de la Terre est un système hautement complexe composé de nombreux compartiments toujours en mouvement et en interaction entre: l'atmosphère, l'hydrosphère, la cryosphère, la surface terrestre et la biosphère qui fonctionne sous l'effet de nombreux facteurs (naturels et anthropiques), à l'origine de sa variabilité et de ses modifications (Figure 2):

Paramètres orbitaux de la Terre: Du fait de l'attraction de la lune et des planètes, la révolution de la Terre autour du soleil n'est pas parfaitement périodique. Ces variations climatiques cycliques sont fonctions de trois paramètres orbitaux : 1) l'excentricité : orbite ellipsoïdale de la Terre avec des pseudo-périodicités autour de 100 ka et 400 ka (Paillard, 2010), 2) l'obliquité de l'axe de la Terre qui oscille entre 21.9 à 24.5° avec des périodicités de 41 ka (Paillard, 2010) et 3) la précession de l'axe de rotation de la Terre avec une périodicité moyenne autour de 19 ka et 23 ka pour ce phénomène) (Laskar et al., 2004, 1993; Paillard, 2010).

Variations de l'activité solaire: L'énergie rayonnée par le soleil varie au cours du temps (Lean et al., 1995). Les mesures réalisées à partir de radiomètres ont permis de mettre en évidence une périodicité autour de 11 ans des fluctuations solaires (Fröhlich, 2006).

Tectonique des plaques et érosion chimique: L'expansion des fonds océaniques déplace les continents et sa vitesse contrôle la proportion de CO₂ délivrée dans l'atmosphère et agirait donc indirectement sur la température de la Terre (Berner, 1999). D'autre part, les événements de la tectonique des plaques (subduction et collision des plaques continentales) agissent sur le relief et influencent la dynamique atmosphérique, les processus d'érosion et donc le climat (Kump et al., 2000; Ruddiman, 2008).

L'effet de serre (naturel): Le système climatique de la Terre est fonction de l'énergie solaire. La moitié de cette énergie (~50%) est absorbée par la surface terrestre. Environ 30 % de cette énergie est réfléchi vers l'espace par les gaz, les aérosols, les

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nuages et la surface terrestre (albédo), tandis que l'atmosphère en absorbe environ 20 %. L'énergie réfléchie par la surface terrestre est en grande partie absorbée par les nuages (qui réémettent aussi ces radiations) et certains constituants de l'atmosphère : vapeur d'eau, dioxyde de carbone (CO_2), méthane (CH_4), protoxyde d'azote (N_2O), et autres gaz à effet de serre...) (Cubash et al., 2013). D'autre part, l'atmosphère principalement composée de gaz, contient aussi des matières liquides et solides sous forme de fines particules (aérosols) qui influencent directement le transfert d'énergie par radiations solaires (absorption et dispersion) et par conséquent le climat et la météo (Boucher et al., 2013). L'effet de serre est donc un processus naturel important agissant sur le climat de la Terre maintenant une température à la surface terrestre propice à la vie, néanmoins sa dynamique peut être impactée par les activités humaines.

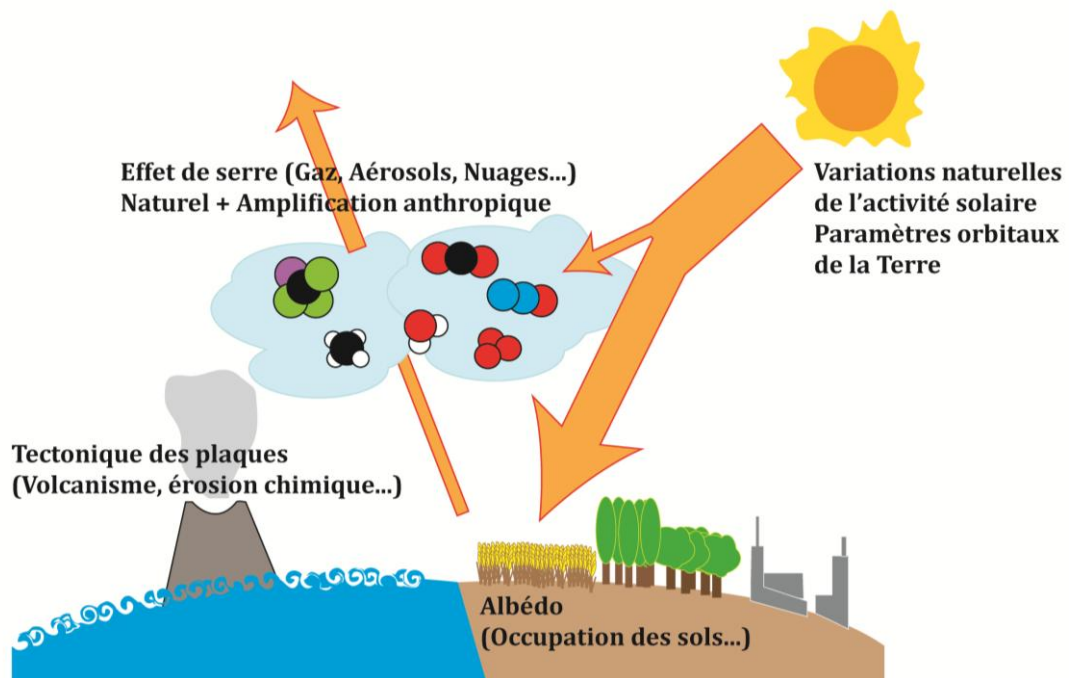


Figure 2. Les principaux facteurs régissant le climat sur Terre

Amplification de l'effet de serre: Un nombre important d'études scientifiques s'est porté sur le fait que la structure naturelle du climat subit des perturbations. Le réchauffement global observé particulièrement sur les deux dernières décennies (augmentation moyenne des températures) est attribué en grande partie aux activités humaines et à l'émission de gaz à effet de serre dans l'atmosphère (Cubash et al., 2013; IPCC, 2014; IPCC, 2007).

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Aérosols: Toutes les activités humaines, telles que l'agriculture et l'industrie, contribuent à importer de grandes quantités d'aérosols dans l'atmosphère. La nature (absorbeurs ou réflecteurs) et la quantité de celles-ci ainsi que la nature de la surface sous-jacente (surface terrestre ou eau) déterminent leur effet positif ou négatif sur le réchauffement (Bellouin et al., 2011; Liao et al., 2009).

Albédo: Enfin l'homme affecte le bilan énergétique de la Terre par son influence sur la couverture de la surface terrestre (anthropisation des terres) en modifiant la manière dont laquelle la surface terrestre redistribue les flux de chaleurs (Foley et al., 2005; Houghton, 2003).

Le climat varie donc sous le contrôle de nombreux facteurs, à la fois naturels et anthropiques (paramètres orbitaux, activité solaire, effet de serre...). En réponse au forçage de ces divers facteurs qui surviennent aléatoirement ou (quasi-)périodiquement, la variabilité du système climatique s'effectue sur toutes les échelles de temps, de l'année au million d'années (Bartlein, 2007; Hugget, 1991; Mitchell, 1976) (Figure 3).

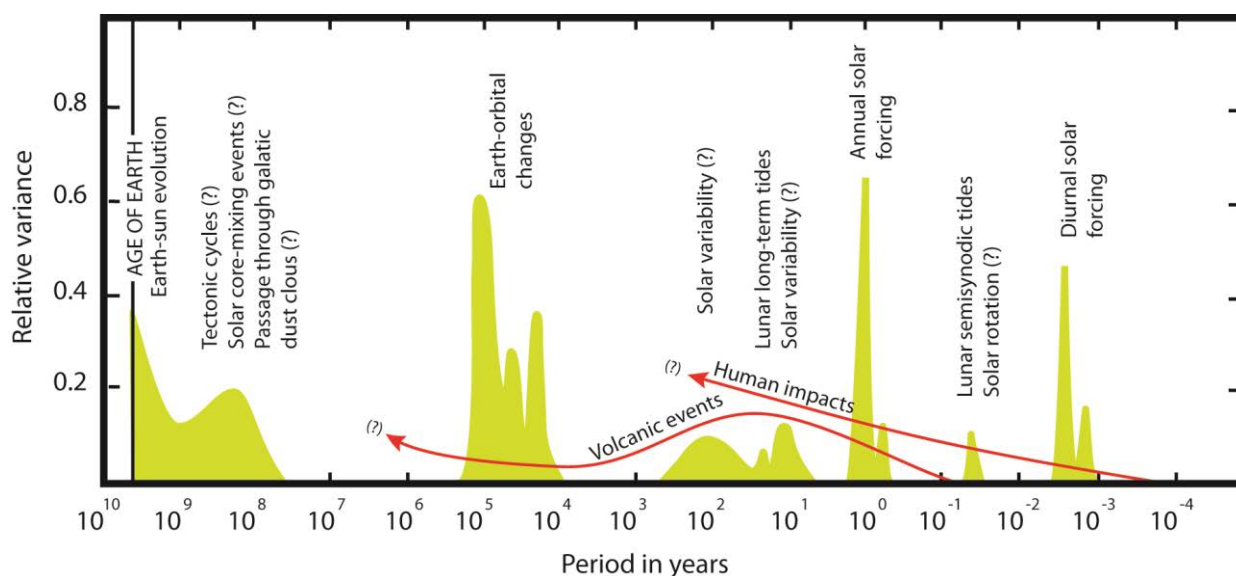


Figure 3. Variation relative des variations climatiques en fonction de la période, modifiée à partir de Mitchell.

Cette variabilité climatique se fait à travers un système hiérarchisé de facteurs de contrôles et de réponses qui apparaissent à toutes les échelles de temps et qui peuvent se traduire à différentes échelles spatiales : globale, hémisphérique, continentale, régionale jusqu'à l'échelle locale par la variation individuelle de variables climatiques (Bartlein, 2007). Les réponses à ces perturbations climatiques peuvent être directes

(hausse ou baisse de température, ...) ou indirectes (eustatisme, ...). Un des exemples les plus récents sont les variations de températures dues à l'alternance des périodes glaciaire/interglaciaire durant le pléistocène. Ces variations de températures sont corrélées à des variations eustatiques importantes (niveau marin à environ -120m correspondant à des températures de -4°C) (Figure 4).

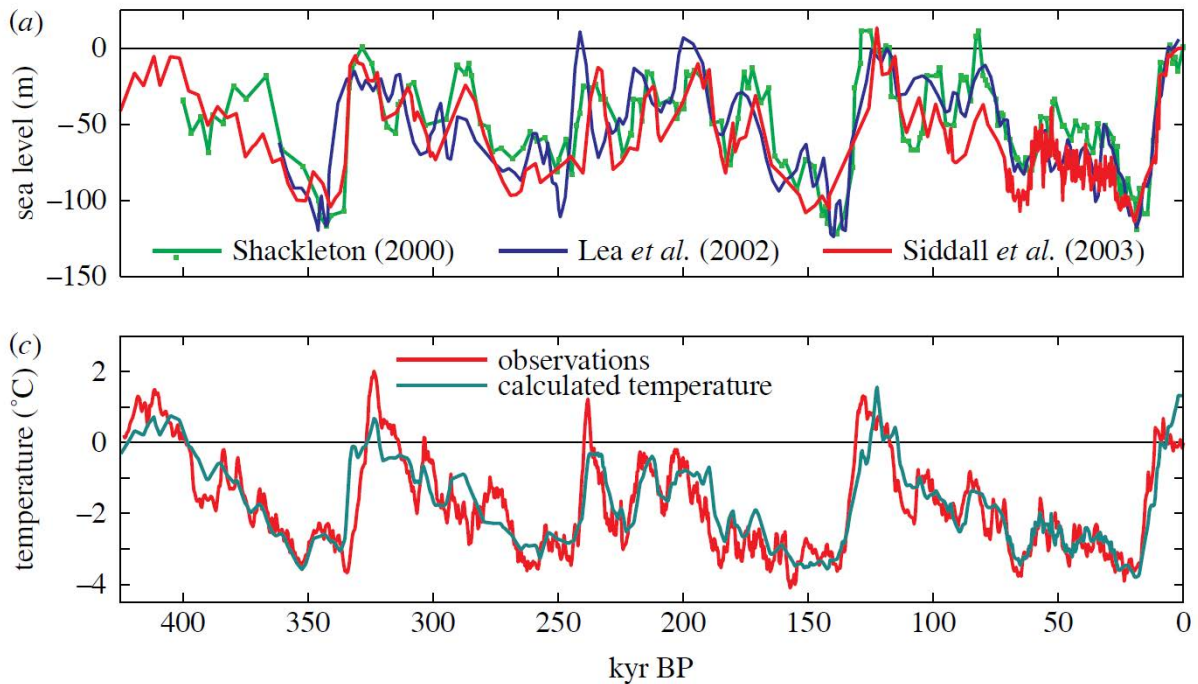


Figure 4. (a) Enregistrements du niveau marin Shackleton (2000; courbe verte), Lea et al. (2002; courbe bleue) and Siddall et al. (2003; courbe rouge). (b) Les variations de température du Paléoclimat: Température calculée et observée (Vostok) extrait de Climate change and trace gases (2007).

C'est pourquoi chaque perturbation d'une variable climatique (température, précipitation...) a le potentiel d'affecter directement ou indirectement le cycle hydrologique et par conséquent la ressource en eau souterraine ainsi que les différents compartiments qui y sont associés (Alavian et al., 2009; Clifton et al., 2010).

1.2. Influence climatique sur la ressource en eau souterraine

Les modifications climatiques interviennent à toutes les échelles de temps et ne sont pas un phénomène nouveau puisque le climat a changé de nombreuses fois en réponse au forçage des divers facteurs. Des liens entre le climat et les eaux souterraines existent donc à toutes les échelles de temps. Allant de réponses à court terme comme les variations de pression atmosphérique et les marées jusqu'à des réponses à long terme comme les variations climatiques à l'échelle de milliers d'années qui ont eu un effet sur la recharge passée et qui ont potentiellement un impact sur le comportement du système actuel. De plus, en comparaison avec les systèmes d'eau de surface, les systèmes souterrains tendent à répondre plus lentement (Kelly et al., 2013; Walton, 2011) aux variations des conditions climatiques. Ces perturbations induisent des changements hydrologiques qui peuvent potentiellement se répercuter sur la ressource en eau souterraine, en relation avec certains mécanismes propres à cette ressource (recharge, décharge, stock) et/ou ses caractéristiques (quantité, qualité).

1.2.1. Influence sur la recharge

La recharge des eaux souterraines peut être définie comme l'eau qui atteint un aquifère que ce soit verticalement ou latéralement (Gerber and Howard, 2000). Naturellement, la recharge des eaux souterraines peut localement avoir lieu à partir des compartiments d'eau de surface et/ou par l'alimentation diffuse des précipitations via la zone non-saturée (Döll and Fiedler, 2008) ainsi que par drainance d'aquifères. La plupart des systèmes hydrogéologiques sont alimentés par les deux types de recharge, mais la proportion de recharge diffuse diminue avec l'aridité de la région (Lemer et al., 1990). Les facteurs influençant la recharge des aquifères peuvent être contrôlés par des forçages externes comme la quantité de précipitation, la température, la vitesse du vent, l'évapotranspiration, l'utilisation du sol et la végétation ou bien contrôlés par les propriétés physiques particulières du milieu comme les propriétés du sol, la géomorphologie, la topographie et la géologie (Jyrkama and Sykes, 2006; van Roosmalen et al., 2007). Néanmoins, les précipitations (efficaces) restent le paramètre climatique moteur de la recharge des systèmes souterrains (Crosbie et al., 2010; Knutssen, 1988). La sensibilité de la recharge aux variations climatiques est reliée aux changements de la

quantité de précipitation effective (précipitations atteignant la partie souterraine) elle-même fortement dépendante de l'évapotranspiration (paramètre lui-même fortement dépendant de la température et donc des concentrations en CO₂) (Barron et al., 2011; Kumar, 2012). Bien que les effets des variations climatiques sur la recharge soient difficiles à apprécier, certains indices conservés dans l'eau souterraine (signatures isotopiques des éléments constitutifs de l'eau et des concentrations en gaz nobles) permettent de contraindre les conditions climatiques au cours de la recharge et la durée depuis laquelle cette recharge s'est effectuée (Bakari et al., 2012), soulignant ainsi que les eaux souterraines sont des archives paléoclimatiques (Edmunds, 2005a) (voir section 2.1.4 du Chapitre 2).

1.2.2. Influence sur la résurgence

La décharge, autre élément clé, est caractérisée par la circulation d'eau au sein d'un aquifère sortante de celui-ci. Les zones naturelles caractéristiques de décharge d'eau souterraine sont les réseaux hydrographiques, les marais, les zones humides, les lacs et les océans. L'intime relation existant entre ces compartiments d'eau de surface et la partie souterraine implique que si un de ces compartiments est impacté par le climat, les autres le seront aussi. Néanmoins, l'impact climatique sur la décharge est moins bien connu du fait du manque de données et de la difficulté à quantifier ces processus (Van Vliet, 2007). En effet, moins d'intérêt a été porté à l'étude des écosystèmes dépendants des eaux souterraines (végétation...) et la décharge des eaux souterraines vers les rivières, les zones humides et les océans. Les interactions des eaux souterraines avec les compartiments de surface sont aussi dépendantes des niveaux relatifs entre eux, l'impact du climat se fait donc ressentir indirectement par l'altération de la recharge et/ou des mécanismes de décharge (comme l'évapotranspiration).

1.2.3. Influence sur le stock (ou emmagasinement)

Du fait de l'affectation des entrées (recharge) et des sorties (décharge) du système hydrologique par les variations climatiques, le stock d'eau au sein du système est lui aussi modifié. Les variations de quantité d'eau (variation de l'emmagasinement) correspondant à la différence entre la recharge et la décharge sont dépendantes des

propriétés du système souterrain et de l'échelle de temps suivant les processus considérés (pouvant aller de la journée aux milliers d'années) (Alley et al., 1999).

Sur le long terme (plusieurs décennies ou plus) et en l'absence de perturbation climatique majeure, le bilan hydrologique d'un aquifère est généralement en équilibre dynamique. D'autre part, ce bilan est aussi soumis aux variations climatiques cycliques, mais se reflètent sur plus court terme. Les tendances des niveaux d'eau sur le long terme reflètent donc les variations du stock d'eau pouvant être la signature d'exploitation de la ressource en eau ou de changement climatique.

Les aquifères peu profonds sont plus sensibles aux variations climatiques se produisant sur des petites échelles de temps (Kundzewicz and Döll, 2009), contrairement aux systèmes profonds qui réagissent aux perturbations climatiques majeures sur de grandes échelles de temps (Clifton et al., 2010). L'influence des variations climatiques sur le stock d'eau dépendra aussi de la durée de renouvellement des eaux souterraines considérées (ressource renouvelable ou « fossile »). Ce concept des eaux souterraines non renouvelables fait référence à la durée de renouvellement des eaux. Dans certaines situations, le laps de temps nécessaire pour renouveler la ressource en eau souterraine peut être très long par rapport à la durée moyenne des activités humaines et aux plans de gestion associés (Margat et al., 2006).

1.2.4. Influence sur la qualité

Enfin, le climat n'affecte pas seulement la recharge, le stock et la décharge des systèmes souterrains, mais peut aussi influencer la qualité de l'eau souterraine (Dragoni and Sukhija, 2008). En effet, la qualité des eaux souterraines dépendante des propriétés physiques, chimiques et biologiques des aquifères est notamment contrôlée par les fluctuations climatiques. La perturbation de la recharge et de la température de l'eau modifie la chimie de l'eau, les communautés bactériennes, le transport de soluté et les temps de résidence (Glassley et al., 2003; Van Vliet, 2007). La salinisation des eaux souterraines constitue l'une des principales causes de dégradation de la qualité de l'eau dans le monde (Kloppmann et al., 2010). L'intrusion d'eau salée peut donc être un problème majeur (Cloutier et al., 2009). Les variations climatiques peuvent entraîner des modifications du niveau marin (expansion des océans, fonte des glaces...) ainsi que des taux de recharge des aquifères et donc se caractériser par des intrusions d'eau de

mer dans les aquifères par modification de l'équilibre existant (Ranjan et al., 2006). La qualité des eaux souterraines et donc leurs caractéristiques chimiques peuvent être des indicateurs de processus particuliers d'interactions entre l'eau et la roche ou d'évènements spécifiques de périodes climatiques passées (Dragoni and Sukhija, 2008).

1.3. Synthèse et problématique générale

La variabilité climatique est caractéristique d'un état altéré du climat. Cette altération du climat s'identifie par un changement dans le comportement du climat moyen et/ou par la variation de ses propriétés qui persistent sur des périodes de temps étendues (de l'ordre de la dizaine d'années à plus). Les origines des variations climatiques sont diverses: processus internes naturels ou forçages externes (comme les paramètres orbitaux) ou encore activités anthropiques. En réponse à ces divers processus, les variations mises en jeu impliquent toutes les échelles de temps et d'espaces.

Les modifications climatiques existent depuis les temps géologiques, les ressources hydrogéologiques ont donc subi ces variations climatiques (comme les périodes glaciaires/interglaciaires) et leurs effets (variation de la recharge, eustatisme...) à plusieurs reprises et à différentes périodes de l'histoire. De plus, le climat actuel que nous connaissons est en train de se modifier et se modifiera inévitablement dans le futur (Dragoni and Sukhija, 2008). Ces caractéristiques climatiques soulignent que les eaux souterraines ont subi des variations climatiques, qu'elles en subissent actuellement les changements et qu'elles en subiront à l'avenir.

Du fait du fort lien entre le climat et le cycle de l'eau, un des aspects les plus importants concernant le changement climatique est son impact sur la ressource en eau souterraine, qui fait partie intégrante de ce cycle et qui est un élément essentiel à la vie et nécessaire dans presque toutes les activités humaines. Dans ce contexte, il devient nécessaire de prendre en compte les circulations souterraines sur toutes les échelles de temps et ainsi de s'intéresser aux phénomènes climatiques passés qui ont ou qui gouvernent son évolution. En effet, l'ajustement des eaux souterraines aux différents forçages (climatiques, géologiques, chimiques...) peut se faire sur des temps géologiques et donc mettre en jeu des phénomènes transitoires. Il a par exemple été mis en

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évidence l'influence de changement climatique à grande échelle, comme les glaciations, sur les écoulements régionaux d'eau souterraine actuels (Lemieux et al., 2008a). L'analyse de la réponse du système et l'identification des perturbations passées à l'origine de ces signaux permettent de comprendre et d'apporter des contraintes sur les écoulements, sur des échelles de temps caractéristiques de ces systèmes. Or, les conséquences sur la ressource en eau souterraine (réponses du système aux perturbations) peuvent se caractériser par des changements en termes de qualité, de quantité et de comportement de la ressource en eau. C'est pourquoi, des études visant à estimer l'impact du changement climatique sur la ressource en eau ont vu le jour ces dernières années (Christensen et al., 2004; Fowler et al., 2007; Wilby et al., 2006).

La problématique de cette thèse s'inscrit dans ce cadre. Elle vise à investiguer les conséquences des variations climatiques sur la ressource en eau souterraine. En effet, dans la conjoncture climatique actuelle et des interrogations qui en résultent vis-à-vis du devenir des ressources hydrogéologiques, il convient de s'interroger sur l'impact des variations climatiques sur les ressources en eau souterraine, comme :

Quels sont les impacts des événements climatiques majeurs passés et leurs conséquences sur les ressources hydrogéologiques ? Question dont les réponses pourraient fournir des informations capitales dans la compréhension du fonctionnement de ces systèmes face aux variations climatiques.

Quels sont et quels seront les impacts des variations climatiques sur les ressources hydrogéologiques en terme de qualité et de quantité ? Mais aussi, comment les pressions anthropiques influencent-elles et influenceront-elles ces ressources en fonction du climat et de son évolution possible ?

Les réponses à ces questions constituent des objectifs essentiels de nos sociétés afin de répondre aux enjeux actuels et futurs concernant les ressources hydrogéologiques (estimation de la durabilité de la ressource, exploitation des ressources énergétiques des systèmes profonds, stockage de déchets radioactifs ou de gaz, etc.). Dans le but de répondre à ces diverses interrogations, cette thèse passe par l'étude de la ressource hydrogéologique et de ces relations avec les compartiments de surface et l'océan afin d'apprécier les effets du climat sur le système hydrologique au sens large. Cette thèse s'articule autour de 3 axes principaux. Il convient de remarquer à ce stade que le choix a

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été fait de répartir l'introduction avec les références appropriées dans le début de chacun de ces 3 points principaux :

Le Chapitre 2 se focalise sur les variations climatiques passées et plus particulièrement aux conséquences d'évènements majeurs (transgression marine et période glaciaire) qui ont affecté la ressource en eau souterraine durant les temps géologiques et qui par leur ampleur ont pu être conservés dans la mémoire du système actuel. L'étude et l'identification de ces évènements (transgressions marines passées) permettent d'apporter des contraintes sur la ressource hydrogéologique à grande échelle de temps et d'espace.

Les deux chapitres suivants visent à investiguer l'impact du climat à travers l'étude des eaux souterraines et de leurs relations particulièrement étroites avec les compartiments de surface et l'océan. Le Chapitre 3 se consacre donc à l'étude de l'impact respectif du changement climatique et des activités anthropiques sur les relations entre l'eau souterraine et les zones humides. Le Chapitre 4 investigate les processus de salinisation des aquifères en s'intéressant aux rivières sujettes à ce jour à l'influence de la mer et qui le seront d'autant plus dans le futur du fait du changement climatique et de l'augmentation du niveau de la mer.

Enfin, le Chapitre 5 présente des perspectives de travaux en termes de modélisation numérique à partir des conclusions du Chapitre 2. Modélisation numérique qui pourrait permettre d'apporter de nouvelles contraintes sur les propriétés hydrauliques du système à grande échelle.

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2.1. Introduction

2.1.1. Dynamique des eaux souterraines

Les matériaux terrestres ayant la capacité à conduire la circulation d'eau peuvent être regroupés en 3 grands groupes distincts de réservoirs, selon leurs propriétés pétrophysiques (J. Bear, 1972):

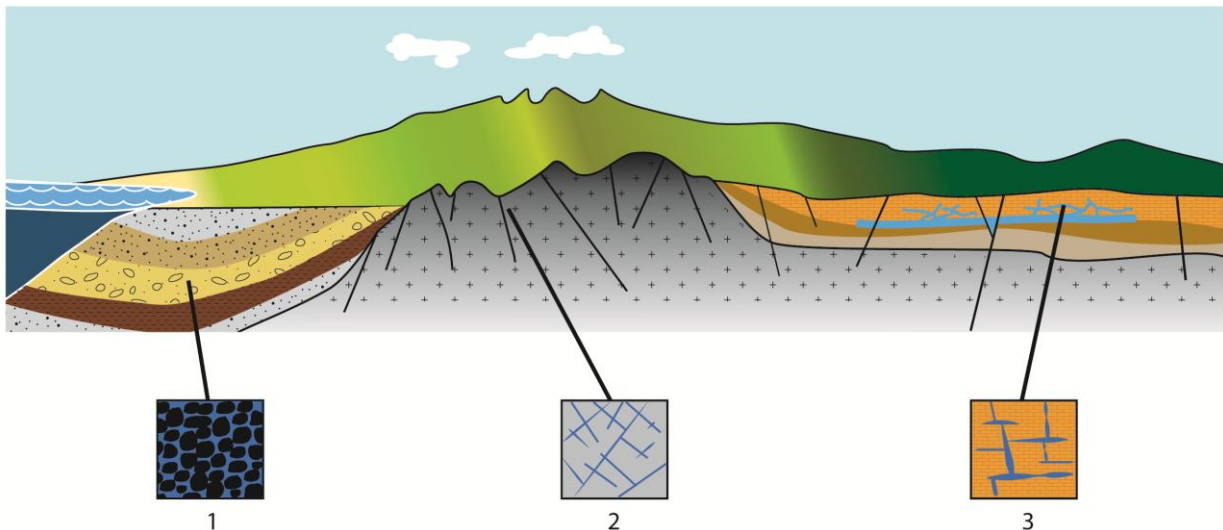


Figure 5. Les grands réservoirs d'eau souterraine, modifiée d'après la figure extraite de l'ouvrage « les eaux souterraines en France ».

- 1- **les aquifères de dépôts sédimentaires non consolidés** (sables, graviers...), caractérisés par une porosité d'interstice originelle créée par les processus géologiques au moment de la mise en place de la roche (Figure 5, n°1).
- 2- **les aquifères des roches fracturées** (cristallines, métamorphiques et volcaniques : granites, gneiss, schistes...), caractérisés par une porosité de fissure ou « secondaire » correspondant à des fissures, des joints, des fractures (Figure 5, n°2).
- 3- **les aquifères des roches sédimentaires solubles** (calcaires, ...), caractérisés par une porosité « secondaire » correspondant à des vides de dissolution (exemple : milieu karstique) (Figure 5, n°3).

La circulation des fluides au sein des roches est donc gouvernée par de nombreux paramètres dont deux principaux: **la porosité (θ)** [sans dimension], ratio entre le volume de vide au sein de la roche et le volume de la roche et la **perméabilité (k)** [m^2] ou plus classiquement la **conductivité hydraulique K** [$m.s^{-1}$] qui caractérise la capacité d'un milieu poreux à se laisser traverser par un fluide. Ces deux propriétés contrôlent le flux des eaux souterraines et donc en partie leur dynamique.

La circulation des eaux souterraines résulte de la différence d'énergie de l'eau d'un point à un autre, l'écoulement se faisant du point avec la plus grande charge hydraulique vers celle de plus faible (h en [m]), charge hydraulique définie par l'équation de Bernoulli :

$$h = z + \frac{p}{\rho \cdot g} + \frac{v^2}{2 \cdot g} \approx z + \frac{p}{\rho \cdot g}$$

avec l'altitude z en [m], la pression de fluide p en [Pa], la densité du fluide ρ en [$kg.m^{-3}$], g l'accélération de la pesanteur g [$m.s^{-2}$] et v [$m.s^{-1}$] la vitesse du fluide qui est typiquement négligée du fait de la faible vitesse de circulation des eaux souterraines (Hubbert, 1940). Le moteur de ces écoulements est donc le gradient hydraulique (différence de charge entre deux points du système sur la distance les séparant), mais la vitesse dépend également des propriétés intrinsèques de l'aquifère (perméabilité, géologie...) ainsi que des propriétés du fluide (Gedeon, 1999).

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Les écoulements souterrains sont décrits par la loi empirique de Darcy, établie en 1856 (Darcy, 1856), loi qui constitue la base de toute analyse quantitative des eaux souterraines:

$$q = \frac{Q}{A} = -K \cdot \frac{dh}{dl}$$

où Q est le débit [$\text{m}^3 \cdot \text{s}^{-1}$], A la section à travers laquelle l'écoulement se fait [m^2], q le flux volumique ou le débit par une unité de surface [$\text{m} \cdot \text{s}^{-1}$], K la conductivité hydraulique [$\text{m} \cdot \text{s}^{-1}$] et $\frac{dh}{dl}$ le gradient hydraulique [sans dimension] qui s'écrit aussi (i). D'autre part, la vitesse moyenne de circulation des eaux souterraines (v) [$\text{m} \cdot \text{s}^{-1}$] peut être estimée à partir de q , en prenant en compte la porosité (θ) (Freeze and Cherry, 1979):

$$v = \frac{q}{\theta}$$

Les écoulements au sein d'un système hydrogéologique résultent de l'équilibre des flux entrants (recharge) et sortants (décharge) entre ces différents composants (compartiments de surface, le milieu géologique et ses limites d'écoulement...). L'équation de continuité permet de décrire cet équilibre pour un fluide incompressible dans un milieu poreux indéformable, à partir de la loi de Darcy (Darcy, 1856):

$$q_x = -K \cdot \frac{dh}{dx}; q_y = -K \cdot \frac{dh}{dy}; q_z = -K \cdot \frac{dh}{dz}$$

et de l'équation de continuité (Bear, 1979):

$$-\nabla \cdot q = S_s \frac{\partial h}{\partial t}$$

L'équation générale d'écoulement des eaux souterraines dans un milieu poreux peut s'écrire :

$$\nabla \cdot (K \cdot \nabla h) = S_s \frac{\partial h}{\partial t}$$

avec t le temps [s], S_s le coefficient d'emmagasinement spécifique [m^{-1}].

2.1.2. Conceptualisation des circulations à l'échelle régionale

La caractérisation des écoulements et la compréhension des milieux hydrogéologiques souvent complexes nécessitent de passer par la conceptualisation. Hubbert (1940) fut le premier à conceptualiser les écoulements souterrains (pour un milieu homogène et isotrope) à partir d'une coupe verticale 2D d'une vallée symétrique (Figure 6).

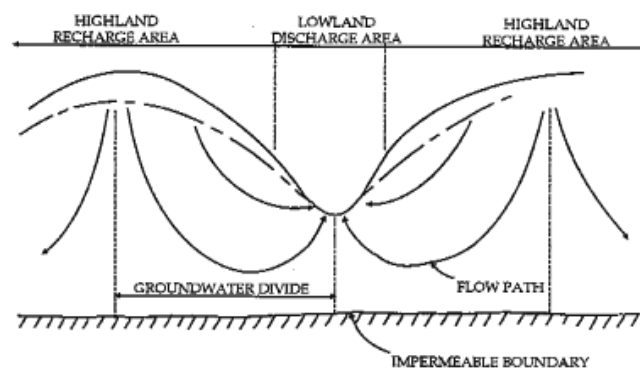


Figure 6. Conceptualisation des écoulements souterrains par Hubbert 1940 pour une vallée symétrique représentée en coupe 2D verticale

Ces travaux permirent de mettre en avant de nombreuses caractéristiques des écoulements souterrains:

- Le niveau de la nappe est un répliquat de la topographie et sa profondeur sous la surface du sol est plus élevée dans les hauts topographiques que dans les bas.
- Les hauts topographiques sont les zones de recharge (écoulements descendants) et les bas topographiques (exemple : vallée) sont les zones de décharge (écoulements ascendants).
- Les écoulements souterrains se font des zones de recharge vers celle de décharge (hauts topographiques vers les bas).
- Il existe des limites « imaginaires » sous les vallées et les crêtes à travers lesquelles il n'y a pas d'écoulement d'eau souterraine (lignes de partage des eaux).

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L'hypothèse de conceptualisation des écoulements à la base de la compréhension de la complexité des circulations (Hubbert, 1940) initia de nombreux travaux théoriques sur les écoulements à grande échelle. Ces études ont en particulier mis en évidence le contrôle de la topographie, de la géologie du milieu (conductivité hydraulique, hétérogénéité...) et du climat sur les écoulements (Freeze and Witherspoon, 1968, 1967, 1966; Toth, 1962), mais aussi l'existence des différentes échelles d'écoulement des eaux souterraines, se répartissant en systèmes locaux, intermédiaires et régionaux (Toth, 1962) (Figure 7).

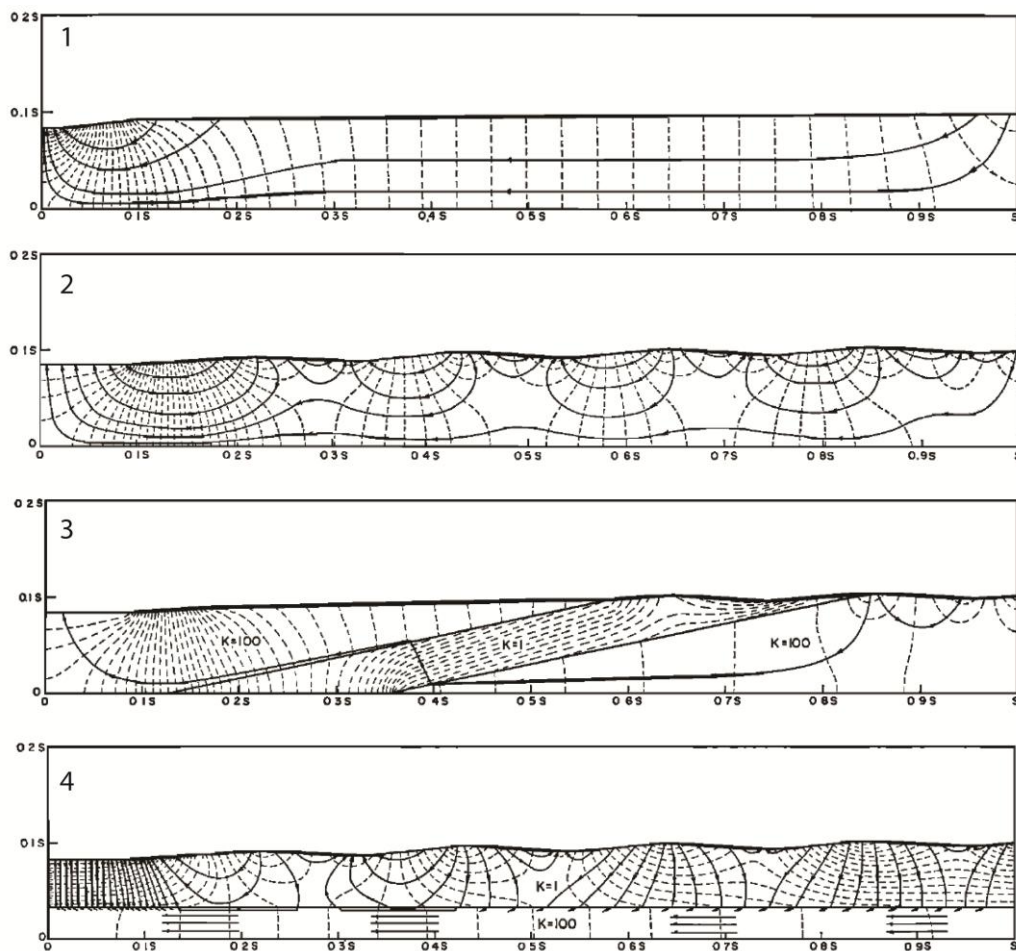


Figure 7. Conceptualisation des écoulements en fonction des propriétés du milieu (Freeze and Witherspoon, 1967; Toth, 1962). 1 et 2 Effets de la configuration du toit de la nappe sur les écoulements à l'échelle régionale à travers un milieu homogène et isotrope. 3 et 4 Effets de la géologie du milieu (conductivité hydraulique et structure géologique) sur les écoulements à l'échelle régionale (Figures extraites de Freeze and Witherspoon, 1967).

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L'extension des systèmes hydrogéologiques peut varier de quelques km² à plus d'une dizaine de milliers de km² (Weeks and Sun, 1986). Les chemins empruntés par l'eau entre la zone de recharge et l'exutoire peuvent donc varier de quelques mètres (système d'écoulement local) à plusieurs centaines de kilomètres (système d'écoulement régional). Ces travaux soulignent l'importance de s'intéresser aux problématiques liées à la ressource en eau à l'échelle régionale. Or, du fait de la complexité des écoulements souterrains à cette échelle, cette problématique reste un sujet de recherche d'actualité (Cardenas, 2007; Craig, 2008; Goderniaux et al., 2013; Jiang et al., 2010, 2009). De récentes études montrent notamment un partitionnement des écoulements avec l'existence de diverses échelles de temps de circulation pour les eaux souterraines (Goderniaux et al., 2013; Jiang et al., 2010).

2.1.3. Échelles de temps des circulations

Sous des conditions naturelles, le temps de résidence (ou temps de trajet) des eaux souterraines de l'entrée du système (zone de recharge) vers un des exutoires (zone de résurgence), correspond au temps nécessaire pour qu'un volume d'eau circule entre ces deux points. L'âge d'un volume d'eau transitant à travers un aquifère pourra s'estimer avec la longueur de la ligne d'écoulement divisée par la vitesse moyenne de l'eau souterraine, la porosité effective du milieu ainsi que par la prise en compte des effets des processus de transports (diffusion, dispersion, mélange, processus d'échange) affectant l'âge de l'eau (Goode, 1996). Si le long de la ligne d'écoulement différentes propriétés hydrauliques sont rencontrées, le temps de trajet total sera la somme des temps de chaque segment (Porter et al., 2008).

Selon les environnements hydrogéologiques, les temps de trajet de l'eau souterraine peuvent varier de moins d'une journée à plus d'un million d'années (Bentley et al., 1986). L'âge de l'eau souterraine (qui augmente constamment le long de la ligne d'écoulement) varie fortement dans les différentes parties du système hydrogéologique. L'eau qui s'infiltre (recharge) à proximité des zones de décharge restera au sein d'un système d'écoulement local et atteindra la zone de décharge en quelques jours ou jusqu'à quelques dizaines années. L'eau s'infiltrant à une distance plus éloignée des zones de décharge suivra un système d'écoulement intermédiaire de profondeur modérée et terminera son parcours au bout de centaines ou de milliers d'années. Enfin,

l'eau qui s'infiltre à côté de la ligne de partage des eaux pourra emprunter une ligne d'écoulement appartenant à un système d'écoulement régional profond et circulera sur des temps géologiques avant d'arriver à la zone de décharge (Figure 8). Néanmoins, ces temps géologiques avec des circulations aussi lentes se retrouvent surtout dans des systèmes aquifères captifs faisant référence à la présence de couches imperméables au sein du système.

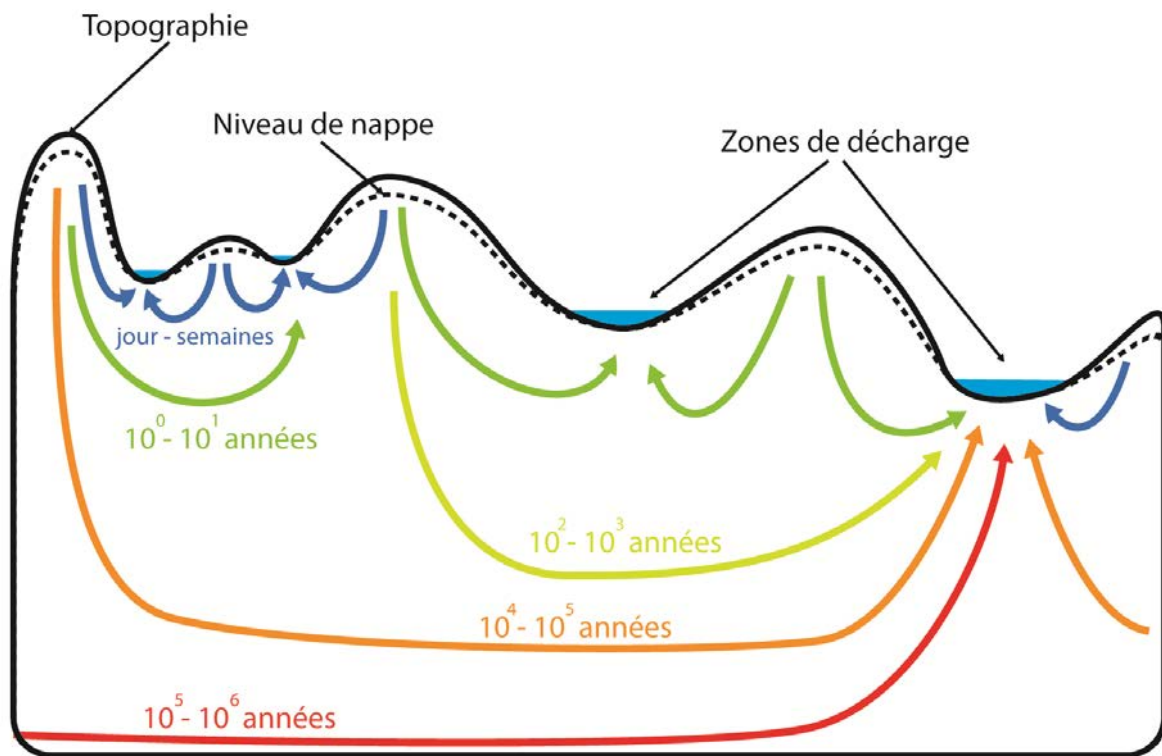


Figure 8. Schématisation des temps d'écoulement des eaux souterraines

Le temps de circulation à travers les systèmes souterrains dépend des variations spatiales et temporelles des gradients de charge hydraulique conduisant les écoulements ainsi que des propriétés du milieu (perméabilité, porosité...) (Alley et al., 2002). Les temps de trajets peuvent donc être influencés par les variations de la recharge (et donc le climat), mais aussi par les activités anthropiques. De plus, les milieux géologiques sont représentés par de larges gammes de perméabilité (avec des variations sur plus de 10 ordres de grandeur) et de porosité (Figure 9 et Tableau 1), d'où une variabilité très importante des temps de circulation des eaux souterraines (De Marsily, 2007; Freeze and Cherry, 1979).

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Tableau 1. Gammes de valeurs de porosité associées à diverses formations géologiques (Freeze and Cherry, 1979)

Formation	θ (%)
Unconsolidated deposits	
Gravel	25 à 40
Sand	25 à 50
Silt	35 à 50
Clay	40 à 70
Rocks	
Fractured basalt	5 à 50
Karst limestone	5 à 50
Sandstone	5 à 30
Limestone, dolomite	0 à 20
Shale	0 à 10
Fractured crystalline rock	0 à 10
Dense crystalline rock	0 à 5

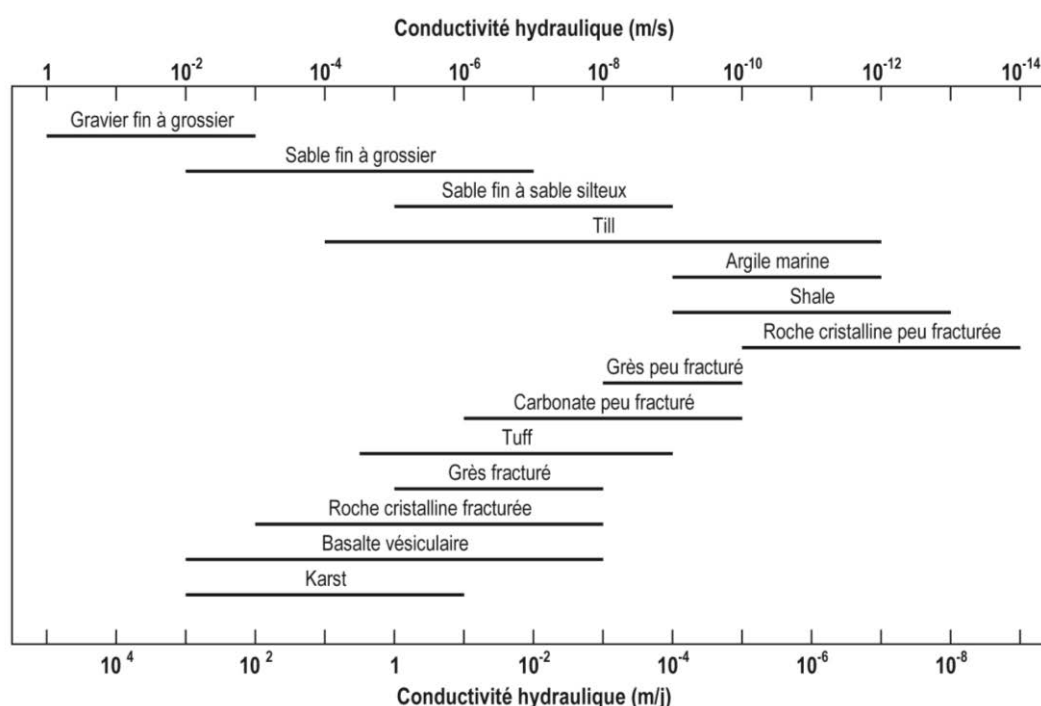


Figure 9. Valeurs de perméabilité et de conductivité hydraulique associées à divers types de roches (d'après Freeze and Cherry, 1979)

Dans le cas d'eaux souterraines appartenant à des systèmes assez lents il convient de s'intéresser aux variations climatiques à long terme ainsi qu'aux changements géologiques (érosion, surrection, tectonique, ... phénomènes qui entraînent des modifications du relief et par conséquent des circulations) pour comprendre les

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circulations d'eau sur plusieurs milliers d'années (Sandford and Buapeng, 1996). Les études de la dynamique de ces systèmes hydrogéologiques passent d'une part par l'utilisation de traceurs environnementaux (^{36}Cl , ^{81}Kr , ^4He , ^3H , ^{14}C , CFC, SF_6 ...), traceurs permettant d'approximer l'« âge » de l'eau sur plusieurs échelles de temps (International Atomic Energy Agency, 2013):

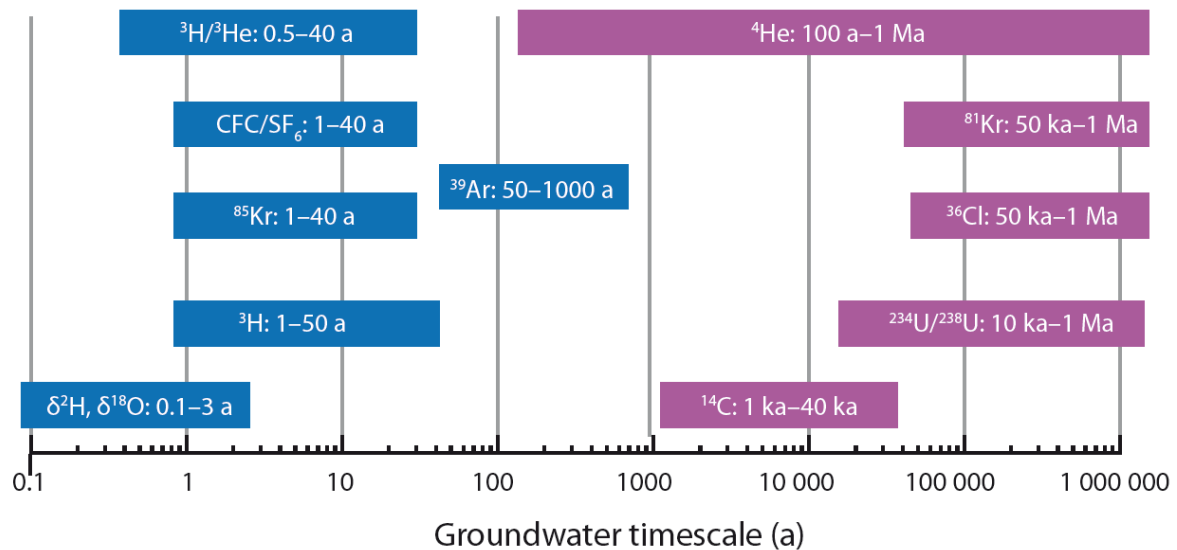


Figure 10. Traceurs chimiques et isotopiques utilisés pour l'estimation de l'âge de l'eau souterraine (International Atomic Energy Agency, 2013).

D'autre part, la compréhension du fonctionnement hydrogéologique peut être appuyée par la modélisation numérique permettant de représenter le système et donc les échelles de temps des écoulements (Ophori, 2004). Or, dans ces modèles des approximations sont nécessaires ce qui peut mettre en doute la fiabilité des résultats. Ces approximations se réfèrent aux incertitudes d'un modèle qui peuvent se distinguer par :

- L'incertitude associée à la paramétrisation du modèle permettant de décrire des processus prenant place sur des échelles temporelles et spatiales qui peuvent être trop fines ou trop larges pour être résolu par le modèle.
- l'incertitude du forçage, relative aux incertitudes des paramètres conducteurs, comme l'incertitude sur l'évolution des émissions de gaz à effet de serre dans les modèles climatiques.
- l'incertitude sur les conditions initiales, l'incertitude et l'inadéquation du modèle.

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Ces diverses incertitudes sur les paramètres clés et les hypothèses (D. Stainforth et al., 2007a; D. A. Stainforth et al., 2007b) se propagent au sein des scénarios issus des modèles climatiques et s'ajoutent aux incertitudes des modèles hydrogéologiques.

C'est pourquoi l'apport d'une contrainte temporelle avec les traceurs permet d'augmenter la représentativité des modèles hydrogéologiques et ainsi de contraindre les échelles de temps de circulation des eaux souterraines, tout en tenant compte des différents effets des processus de transport (Bentley et al., 1986; Goode, 1996; International Atomic Energy Agency, 2013).

2.1.4. Les eaux souterraines archives du climat

Les eaux souterraines ont été reconnues comme étant une archive des conditions climatiques passées et des changements hydrologiques (Edmunds, 2005). Depuis cette mise en évidence de l'effet des changements climatiques sur la disponibilité en eau, un fascinant défi consistant à relier les enregistrements hydrologiques à des périodes de l'histoire est apparu. La composition chimique et isotopique des eaux souterraines est influencée par les conditions climatiques au moment de la recharge. Ainsi, elles constituent des indicateurs de changement climatique (Fontes et al., 1993). Ces informations stockées sont enregistrées par différents traceurs environnementaux (gaz nobles : Ne, Ar, Kr, Xe... ; isotopes stables de l'eau : ^2H et ^{18}O ; tritium ; radiocarbone ; etc....) qui intègrent différents signaux comme :

- l'origine des précipitations (rapport $^2\text{H}/^1\text{H}$ et $^{18}\text{O}/^{16}\text{O}$ qui varie en fonction de la distance à la mer, de l'altitude ainsi que de la température) (Clark and Fritz, 1997; Clark et al., 1997; Currell et al., 2010).
- la température de recharge et donc les paléotempératures de recharge. La solubilité des gaz (gaz nobles) est fonction de la température ce qui impacte sa concentration dans l'eau (Aeschbach-Hertig et al., 2002, 2000; Beyerle et al., 1998; Stute and Schlosser, 1993). Ce processus est décrit par la loi de Henry qui régit le comportement des gaz mis en contact avec les liquides. La quantité de gaz dissous sera fonction: de la pression partielle du gaz, du coefficient de solubilité du gaz (constant pour un gaz donné dans un liquide donné), de l'inverse de la température et de la durée du contact gaz / liquide.

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- les temps de résidence des eaux souterraines avec des traceurs issus d'activités industrielles ou naturelles marquant une période précise (CFCs, SF₆, ³H et ⁸⁵Kr (Ayraud, 2005; Katz, 2004; Labasque, 2013)) ou accumulatifs comme l'hélium, radioactifs (¹⁴C, ³H, ³He, ³⁹Ar, ³⁶Cl...) (Currell et al., 2010; Raidla et al., 2012; Stute and Deak, 1989; Stute et al., 1995).
- les paléodynamiques atmosphériques, les taux et les sources de recharge par mise en relation de différents traceurs (Gonçalvès et al., 2013; Kreuzer et al., 2009).
- ainsi que le mélange d'eaux au sein du système hydrogéologique (hydrochimie et isotopie) (Beyerle et al., 1998; Douglas et al., 2000; Rueedi et al., 2005).

Les traceurs environnementaux et isotopiques sont donc logiquement devenus au cours du temps des outils fondamentaux dans l'évaluation de la ressource en eau souterraine (par l'apport de contraintes sur la dynamique des écoulements). Néanmoins, du fait de la perturbation du signal climatique au sein du système par des processus diffusifs-dispersifs (Davidson and Airey, 1982) et/ou d'interactions eau-roche (Clark and Fritz, 1997) ces informations paléoclimatiques issues des eaux souterraines sont de faible résolution (+/- 1000 ans) (Stute and Schlosser, 1993). Les incertitudes sur les datations, comme dans le cas du ¹⁴C, découlent de l'incertitude concernant l'activité du ¹⁴C de l'eau à l'état initial ainsi que des différents bilans de masse pour un échantillon. Les modèles sont donc indispensables pour déterminer les incertitudes reliées à l'état initial de l'eau et permettent d'obtenir une gamme d'âges possible. Malgré cette limite de résolution, les signaux chimiques (concentration ou gradients de concentration d'élément chimique) et isotopiques (gaz nobles, isotopes de l'eau, ...) témoignent des événements climatiques majeurs et informent sur la réponse des systèmes hydrogéologiques à ces forçages climatiques.

De ce fait, les réponses à long terme de systèmes hydrogéologiques au forçage climatique ont pu être détectées à partir d'évidence paléohydrologique à l'échelle d'aquifères régionaux (de Vries et al., 2000; Edmunds et al., 2003; Lehmann et al., 2003; McMahon et al., 2004; Patterson et al., 2005). L'emploi de ces traceurs environnementaux dans des régions arides ou semi-arides appuyés par d'autres techniques (comme la modélisation numérique...) a permis de reconstruire l'histoire des

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eaux souterraines dans différentes régions du globe (Afrique du Sud et du Nord, USA, Australie) et de mettre en évidence des paléorecharges de plusieurs milliers d'années, voire jusqu'au million d'années à l'aide de la datation au ^{36}Cl (Scanlon et al., 2006). Dans les régions humides, les systèmes de faible profondeur contiennent généralement des eaux souterraines « récentes », caractérisées par exemple par la présence de ^3H (âge inférieure à 50ans). Néanmoins si des eaux anciennes persistent aujourd'hui dans ces systèmes sous climat humide c'est habituellement le résultat de leur isolation du système actif d'écoulement du fait d'une certaine profondeur ou de conditions hydrauliques limitantes (Jim Hendry et al., 1991). La conservation de ces eaux souterraines anciennes, d'origine météorique, nécessite un écoulement qui a permis à une fraction de ces eaux d'atteindre une profondeur suffisante pour être isolée du système d'écoulement actif sur des échelles de temps de plusieurs milliers à plusieurs millions d'années. Ces conditions peuvent être remplies dans le cas de grands systèmes aquifères, d'aquifères captifs et artésiens, un des exemples le plus connus est le « Great Artesian Basin » en Australie (Torgersen, 1991).

Les cartes piézométriques actuelles permettent de définir les écoulements souterrains récents, mais ne peuvent être directement utilisées pour comprendre les paléocirculations. Néanmoins, la cartographie de la composition chimique et isotopique des eaux souterraines peut constituer une alternative intéressante pour contraindre et comprendre les paléoécoulements. Les différences entre le système d'écoulement actuel et celui suggéré par les traceurs permettent de mettre en évidence des changements des conditions hydrauliques (Plummer et al., 2004; Sanford et al., 2004). Concernant les traceurs réactifs, des réactions géochimiques peuvent avoir lieu le long des lignes d'écoulements, la composition chimique de l'eau évolue donc avec la direction de l'écoulement. Dans ce cas, la cartographie des faciès hydrochimiques (Back, 1966) à grande échelle peut permettre de contraindre les écoulements à long terme, du fait de l'alignement préférentiel de ces faciès perpendiculairement à la direction d'écoulement (Glynn and Plummer, 2005). D'autre part, les traceurs non réactifs (gaz nobles, isotopes de l'eau, chlorure, bromure, sodium, lithium, etc. ...) peuvent fournir des informations sur les directions d'écoulements (du fait de leur distribution qui se fait parallèlement à la direction de circulation), ainsi que sur les différentes origines des eaux dans le système souterrain et les temps de résidence (Glynn and Plummer, 2005).

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Les données géochimiques (concentrations, gaz et éléments dissous, valeurs isotopiques...) dans les eaux souterraines peuvent donc être utilisées pour tracer les circulations de fluides sur l'échelle de temps de circulation du système hydrogéologique. Les travaux qui suivent s'inscrivent dans cette démarche.

L'objectif de la première étude est d'apporter des contraintes sur les temps de circulations de fluide dans les aquifères continentaux à l'échelle du Massif Armoricaire par l'utilisation de traceurs environnementaux inertes (chlorure, bromure). L'analyse de la distribution spatiale des concentrations en chlorures à l'échelle régionale (Massif Armoricaire) au sein des aquifères est utilisée dans le but de contraindre l'hydrodynamique des systèmes hydrogéologiques du Massif Armoricaire à grande échelle spatiale et temporelle.

L'objectif de la seconde étude est de compléter l'histoire hydrogéologique des aquifères du Massif Armoricaire. À partir, du jeu de données de la précédente étude et d'un jeu de données complémentaires regroupant des données géochimiques (sulfate, bromure et chlorure), isotopiques (bore, soufre et oxygène des sulfates), et les gaz nobles dissous (Ne, Ar) de nouvelles contraintes sont apportées sur le fonctionnement hydrodynamique à l'échelle régionale.

Le premier article présenté dans la section 2.2 s'organise comme suit : après une introduction (section 1), nous présentons le contexte géologique (section 2) et le contexte hydrogéologique (section 3) du Massif Armoricaire, la base de données relative au traceur non-réactif (chlorures) utilisée dans cette étude (section 4) ainsi que les derniers événements marins majeurs ayant pu affecter la région d'étude (section 5). La section 6 quant à elle a pour objectif premier de présenter les résultats obtenus vis-à-vis de l'origine du traceur (section 6.1 et 6.2) et dans un deuxième temps de proposer une interprétation de ceux-ci visant à contraindre temporellement les écoulements (section 6.3). La section 7 constitue la discussion des résultats : avec une ouverture comparative sur les distributions de ce traceur répertoriées dans d'autres régions du monde (section 7.1), sur la contrainte temporelle proposée (7.2) et sur l'apport de cette contrainte pour la conceptualisation du fonctionnement hydrodynamique des aquifères armoricains (7.3).

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Le deuxième article présenté dans la section 2.3 qui complète le précédent s'organise comme suit : après une introduction sur le contexte global de l'étude, les contextes géologique et hydrogéologique sont rappelés (section 1). La section 2 est consacrée à la présentation des résultats concernant l'origine du soluté (section 2.1), la distribution verticale des chlorures (section 2.2), la température de recharge (section 2.3) et la datation de l'eau souterraine (section 2.4). La section 3 discute la signification de ces résultats afin d'apporter des contraintes supplémentaires sur l'histoire hydrogéologique des aquifères du Massif Armoricaïn.

Mon rôle au sein de ce dernier travail synthétisé sous forme d'un article récemment soumis au journal PNAS (Proceedings of the National Academy of Sciences) a été principalement centré sur les travaux de la signature marine passée et peut se définir en plusieurs points :

- 1- Travaux sur la signature marine au sein des aquifères du Massif Armoricaïn à l'échelle régionale (voir section 2.2).
- 2- Identification des sites ayant une concentration élevée en chlorures et pouvant permettre un accès afin de réaliser des prélèvements complémentaires dans le but d'enrichir notre base de données (sites complémentaires avec datation, Gaz Noble...).
- 3- Organisation des missions de terrain et réalisation des prélèvements d'eau souterraine sur sites (Datation, Chimie, Gaz Noble...).
- 4- Mesures de Gaz Noble et traitement de données permettant l'identification de la température de recharge associée.
- 5- Enrichissement de la base de données générale (chimie, etc.).
- 6- Bibliographie afin de cerner la période glaciaire caractéristique.

2.2. *Time scales of regional circulation of saline fluids in continental crystalline rock aquifers (Armorican massif, Western France).*

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Time scales of regional circulation of saline fluids in continental aquifers (Armorican massif, Western France)

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Time scales of regional circulation of saline fluids
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ABSTRACT

In recent decades, saline fluids have been sampled worldwide at great depths in continental basements. Although some of them have been attributed to marine transgressions the mechanisms allowing their circulation is not understood. In this paper, we describe the horizontal and vertical distribution of moderately saline fluids (60 to 1400mg/l) sampled at depths ranging from 41m to 200m in crystalline rock aquifers at the regional scale of the Armorican Massif (northwestern France). The horizontal and vertical distributions of high chloride concentrations are in good agreement with both the altitudinal and vertical limits and succession of the three major transgressions between the Mio-Pliocene and Pleistocene ages. The mean chloride concentration for each transgression area is exponentially related to the time spanned until present. It defines the potential laws of leaching (displacement) of marine waters by fresh meteoric waters. The results of the Armorican aquifers provide the first observed constraints for the time scales of seawater circulation in the continental crystalline basement and the subsequent leaching by fresh meteoric waters. The general trend of increasing chloride concentration with depth and the time frame for the flushing process provide useful information to develop conceptual models of the paleo-functionning of Armorican aquifers.

Keywords: Saline fluids, crystalline basement, groundwaters, paleohydrogeology, geologic time scale, climatic events.

Highlights:

- The Armorican massif presents clear evidences of a marine origin of the saline component in the fluids at the regional scale.
- High chloride concentrations are attributed to three past marine transgressions.
- Saline fluids provide constraints for the time scales of fluid circulation.
- The general trend of increasing chloride concentration with depth supports the seawater introduction by gravity-driven flow at depth in the basement.
- The time frame for the flushing process is useful information to develop conceptual models of the paleo-functioning of Armorican aquifers.

1. INTRODUCTION

In recent decades, saline fluids have been sampled at great depths (0.5 - 5 km) in continental basements (Bucher and Stober, 2010; Frape et al., 2003). In several places, these fluids have been considered as old seawater introduced in aquifers during marine transgressions (Aquilina and Dreuzzy, 2011; Beaucaire et al., 1999; Bottomley et al., 1999, 1994; Douglas et al., 2000; Greene et al., 2008; Louvat et al., 1999; Négrel and Casanova, 2005). Two alternative origins related either to glacial recharge of brines due to cryogenic mechanisms (Starinsky and Katz, 2003), or to anthropogenic sources (Kelly et al., 2008; Mullaney et al., 2009; Panno et al., 2006; Perera et al., 2013) have also been proposed. These three hypotheses have led to extremely different estimates of the saline fluid residence time (Devonian or Pleistocene for the Canadian brines, for example) and thus of the paleohydrogeology of the continental aquifers. Although thorough investigations have been carried out in several places, the origin and fate of the salinity are still issues. We have almost no field observations identifying the potential mechanisms leading to marine fluid introduction and conservation in hard rock aquifers, as well as the influence of glacial recharge at various depths in these aquifers remain. Furthermore, the time-scales of these mechanisms are unknown.

In this study, we investigated the effects of marine transgressions in the spatial (both horizontal and vertical) distribution of chloride concentrations at a regional scale. Since the transgression characteristics differ from one event to another, we expect to find higher concentrations in the areas flooded by the later. The decrease of chloride concentrations with time potentially provides constraints for fluid circulation time-scales in the basement. This concept is based on the fact that during transgression, a large amount of chlorides was introduced in flooded areas. Secondly, the chloride concentration of the submerged area decreases under the influence of dilution/leaching by meteoric waters. Thus, the decline in chloride concentration since the time of the last transgression is characteristic of the dilution/leaching efficiency. This makes it important to identify the last transgression that has flooded the considered area.

The study focuses on the Armorican basement (western France), where several authors have reported evidence of saline fluids (Ayraud et al., 2008; Pauwels et al., 2010). Three main transgressions were recorded during Mio-Pliocene to Pleistocene

times, the latest being the highest. This is a particularly favorable context since a large part of the area flooded by the oldest and highest transgression remains unaffected by later transgression events.

2. GEOLOGICAL BACKGROUND

The Armorican basement extends over an area of 68,500 km² in the northwestern part of France. The Armorican massif is a crystalline basement localized between the English Channel and the Bay of Biscay. The northern part corresponds to the Cadomian orogenic belt (Chantraine et al., 2001). The central and southern zones belong to the European Variscan belt (Le Corre et al., 1991). The Armorican massif is a basement made of Upper Proterozoic to Paleozoic formations mainly composed of low and high metamorphic rocks (schists, sandstones and gneisses...), plutonic (granite) and volcanic (basalt) rocks deformed during the two orogenies (Cadomian and Variscan) (Figure 11). These orogenies have initially developed the main tectonic structures of the Armorican massif: the North and the South Armorican Shear Zones (NASZ and SASZ, respectively), and the Quessoy/Nort-sur-Erdre Fault (Bonnet et al., 2000) (Figure 11). These structures have been reactivated during Mesozoic and Cenozoic periods. Tertiary and quaternary geodynamic evolution of the Armorican massif are related to the collision between Europe and Africa (Brault, 2002; Gros and Limasset, 1984; Ziegler, 1990). This evolution mainly consists in three major steps (Thomas, 1999): (1) From the end of Cretaceous to the upper Eocene, the Armorican massif is subjected to a lithospheric buckling due to the Pyrenean compression. (2) From the upper Eocene to Oligocene the buckling decreases and the geodynamic evolution is driven by a generalized extension. (3) From the Mio-Pliocene to present days, the Armorican massif is uplifted due to a lithospheric buckling in relation with the Alpine compression. This compression is still active as observed through the Armorican massif seismic activity (Lenôtre et al., 1999; Nicolas et al., 1990).

Local topography is controlled by rock lithologies and, on a larger scale, by tectonic faults (Northern and southern shear zones (Dugué, 2007; Gumiaux et al., 2004)). It is relatively moderate with three domains of higher elevation above 200m and a maximum elevation of 416m in the east (Figure 12).

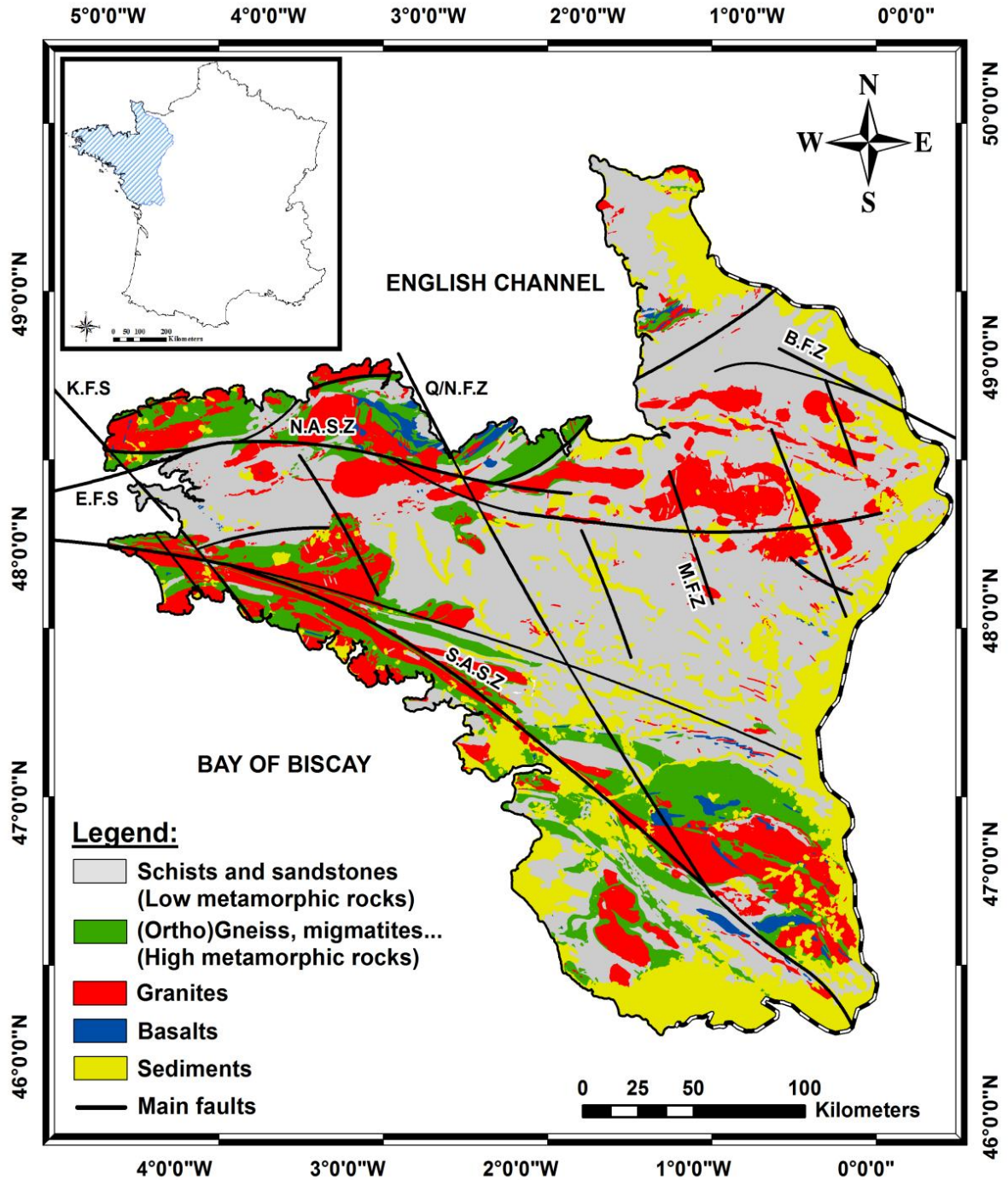


Figure 11. Simplified geological map of the main lithological units and main geological structures of the Armorican Massif: NASZ: North American Shear Zone; SASZ: South Armoricain Shear Zone; Q/NFZ: Quessoy/Nort-sur-Erdre Fault Zone; MFS: Mayenne fault System; KFS: Kerforne Fault System and EFS: Elorne Fault system (according to Bonnet et al. 2000).

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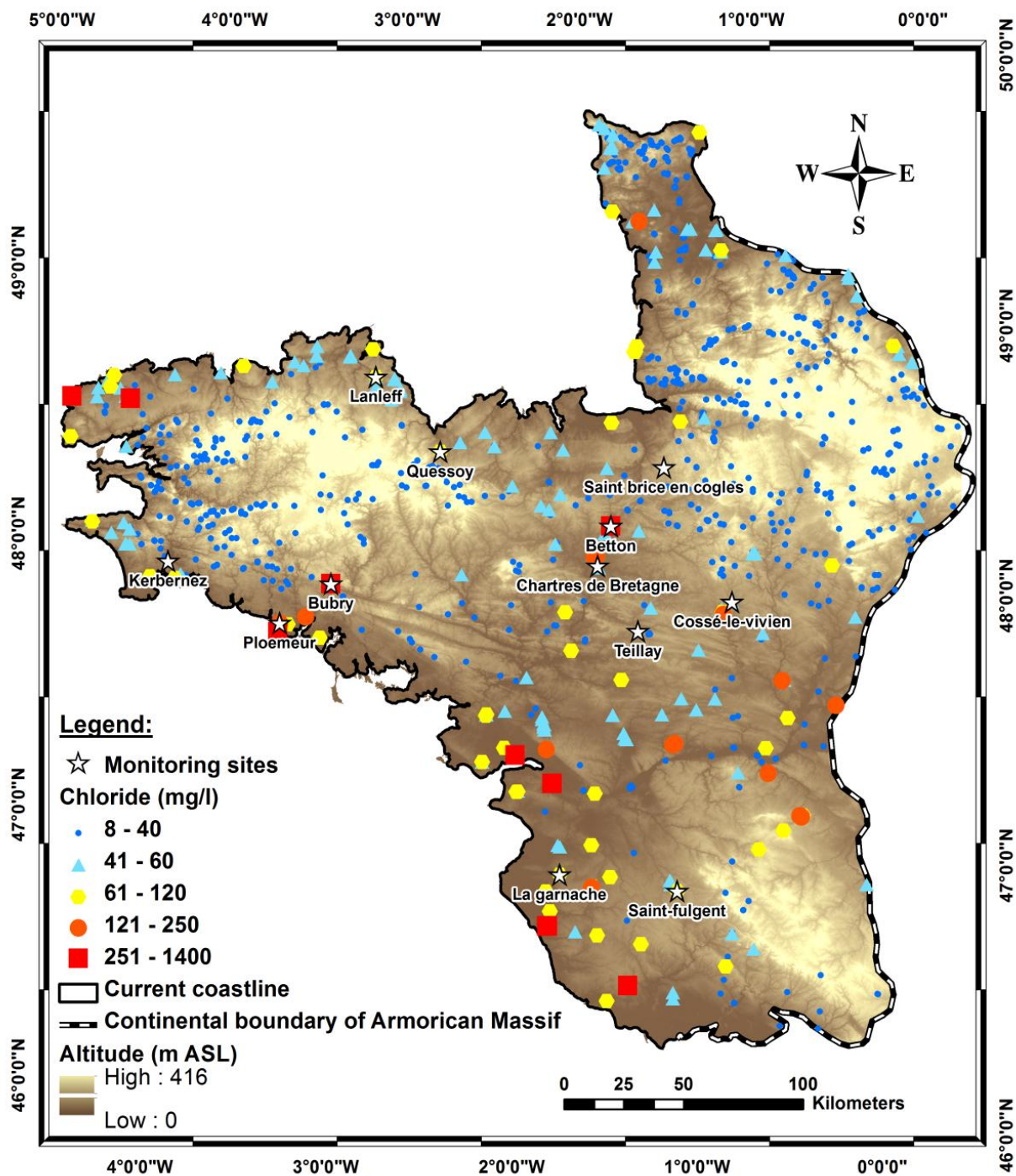


Figure 12. Map of the Armorican Massif including the distribution of chloride concentration for the whole area (from preprocessed chloride database) and the location of the 12 sites investigated.

3. HYDROGEOLOGICAL SETTING

The main groundwater flow in crystalline rock aquifers is considered to be localized in superficial formations resulting mainly from rock weathering, and below in fracture and major geologic discontinuities networks (Larsson, 1987; Stober and Bucher, 2007; Wyns et al., 2004). The compartmentalized structure includes various reservoir scales and characteristic physical parameters (permeability, porosity...) varying over several orders of magnitude (Clauser, 1992). At the scale of the Armorican massif a weathered layer 20 to 30m thick is present in many places, and likely ubiquitously (Brault, 2002; Dewandel et al., 2006). Roques (2013) has studied the influence of fault zones for sites characterized by high groundwater yields at the scale of the Armorican massif. At this scale, groundwater resources in the shallower part (<20m) are widespread but limited in term of productivity. Conversely, high groundwater productivities are associated to fault zones below the weathered zone. Indeed, groundwater resources at significant depth have been found in the Armorican bedrock (Le Borgne et al., 2006; Leray et al., 2013; Roques et al., 2014) and it can be considered that the fractured system constitutes a water reservoir in the continental basement. Free groundwater circulation in crystalline environments has been reported from several studies (Banks et al., 1996; Larsson, 1987). Several studies have shown that basement is generally characterized by an interconnected fracture system at several km depth (Aquilina and Dreuzzy, 2011; Edmunds and Savage, 1991; Stober et al., 2002). Hydrodynamic properties of crystalline rock aquifers of the Armorican basement have been intensively investigated in several research sites (Ploemeur and St-Brice-en-Cogles sites, Figure 12) (Dorn et al., 2012; Le Borgne et al., 2006, 2004; Leray et al., 2013; Roques, 2013; Roques et al., 2014; Touchard, 1999) (Figure 12). Moreover, previous studies in the Armorican basement have identified a compartmentalization of these crystalline rock aquifers from hydrological, geochemical, geological and geophysical data (Ayraud et al., 2008; Roques, 2013; Roques et al., 2014), and from regional-scale numerical modeling (Goderniaux et al., 2013). Geochemical studies also showed specific chemical compositions of the water in the deep fractured aquifer, potentially including saline groundwaters (Aquilina et al., 2013; Ayraud et al., 2008; Pauwels et al., 2013).

4. CHLORIDE DATABASE

The quantity supplied and the quality of the main groundwater resources in the Armorican massif are monitored through a public well network (ADES: “banque nationale d’Accès aux Données sur les Eaux Souterraines”, National database for groundwater resources, <http://www.adeseaufrance.fr/>). For most of these wells hydrogeological informations (geographical location, altitude, depth, drilling log, borehole parameters, water inflow points, etc...) are available through a second public database of drillings (BSS: “Base de données du Sous-Sol”, <http://infoterre.brgm.fr/>). The precise location of water inflows is not described for each well, and it has been considered that the water inflow corresponds to the base of the well. This assumption is valid taking into account the drilling goal which is to stop the well drilling when a sufficient water inflow is found.

The network of wells investigated covers the whole Armorican basement and includes more than 1800 drillings with an average depth of approximately 40m. Chemical records, such as chloride or nitrate concentrations, are available for each drilling. We first preprocessed the database of 1874 chloride measurements and removed wells that are potentially not representative of natural chloride concentrations. We removed wells close to a potential anthropic pollution source (industrial estate, garbage dump, mill pond...), and wells with a large variability through time (large standard deviation of chloride concentration higher than 30%). We also did not consider the wells at a distance less than one kilometer from the current coastline, whose concentration may be directly influenced by current saltwater intrusion. 716 of the 1874 wells have been excluded from the database, i.e. 38% of the total. The vertical distribution of the chloride concentrations for the remaining values was analyzed by intervals of 10m from an altitude of 350m above sea level to -125m. The average chloride concentration and the 90th percentile were calculated for each 10m interval.

5. MARINE TRANSGRESSION CHARACTERISTICS

Chlorides of marine origin that are not brought by rainfall (i.e. concentrations higher than 40 mg.l⁻¹) are likely related to marine transgressions that flooded the Armorican massif from times to times. The likely scenario is that a fairly large amount of chlorides is injected during each transgression in submerged regions, and that the salt concentration decreases from the end of the transgression event due to dilution or leaching by meteoric waters. If the leaching/dilution is efficient enough, for instance if the time lag between two successive transgressions is larger than the leaching time, the system is fully reset by the last event (i.e. the amount of chloride injected during the last event is much larger than the residual of formers). The objective of this part is to characterize the last transgressions that have flooded the Armorican massif.

According to stratigraphic record studies in the Armorican Massif and, to the eustatic sea-level fluctuations (Figure 13 ; Hardenbol et al., 1998), three main transgressions can be identified since the Mio-Pliocene. The oldest of Messinian age has been identified in several places (Néraudeau et al., 2003; Van Vliet-Lanoe et al., 1998), and dated between 6 and 4.6 Myr by several methods (ESR: Electron Spin Resonance Spectroscopy on quartz, ⁸⁷Sr/⁸⁶Sr on shells and bones in “Redonian” shelly sands or “Faluns” (Mercier et al., 2000; Néraudeau et al., 2002). These sediments would attest of the maximum transgression of the late Neogene in northwestern France (Néraudeau et al., 2010). This transgression corresponds to a high sea level of +90m according to Hardenbol et al., (1998). The second transgression is mostly characterized by the deposition of clays, which contains few bioclastic constituents (Morzadec-Kerfourn, 1997, 1977). The so-called Redon clays formation are dated as Pliocene: Piacenzian (Reuverian, ~2.7+/-0.3 Myr) from a pollen analysis (Morzadec-Kerfourn, 1997, 1982). The corresponding sea level is around +60m according to the chart (Hardenbol et al., 1998). The last known transgression in the area is characterized by clay deposits that cover the Redon clays (Morzadec-Kerfourn, 1997). According to pollen analysis of the Lanrinou clay at Landerneau (Morzadec-Kerfourn, 1997, 1982), these sediments can be correlated with the lower Pleistocene around 1.6-2 Myr (Gelasian/Calabrian). The corresponding event coincide with the high sea-level stand of +30m (Hardenbol et al., 1998).

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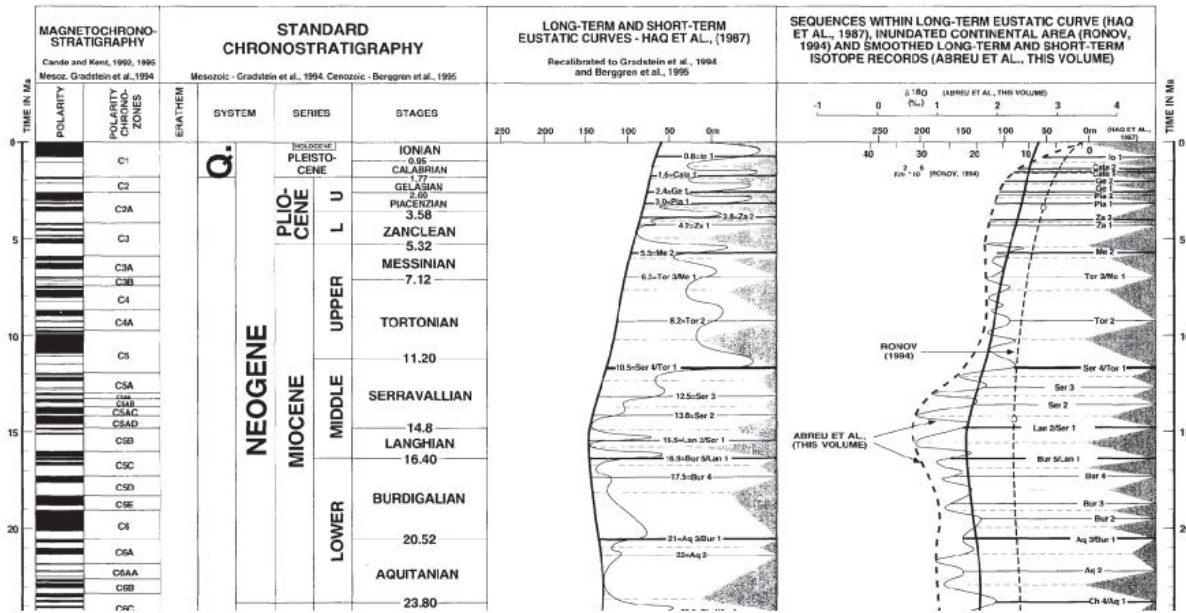


Figure 13. Extract from Mesozoic and Cenozoic sequence chronostratigraphic chart established by Hardenbol et al. 1998.

Thus, at least three marine transgressions, with three different paleocoastlines, must be considered for the Armorican massif: the oldest, which is also the highest, is Mio-Pliocene: Messinian ($\sim 5.3 \pm 0.8$ Myr) with sea level +90m asl, the second is Pliocene: Reuverian ($\sim 2.7 \pm 0.3$ Myr) with sea level +60m asl; and the most recent is Pleistocene: Gelasian/Calabrian ($\sim 1.8 \pm 0.2$ Myr) with sea level +30m asl.

Using local studies on the large-scale relief development and the paleotopographic evolution of the Armorican basement (Bonnet et al., 2000, 1998; Brault et al., 2004; Guillocheau et al., 2003; Lague et al., 2000; Lenôtre et al., 1999; Morzadec-Kerfourn, 1997; Van Vliet-Lanoe et al., 1998), a paleotopography was reconstructed for the whole area.

Globally, the topography evolution is the result of tectonic movements, climate and eustatic variations since the Mio-Pliocene. The modern topography is characterized by incised valleys with an incision average depth of 60-100m and significant topographic variations. The onset of the current incised and dense hydrological network is probably not older than 1 Myr (Bonnet et al., 2000, 1998; Lefebvre et al., 1994). This topography contrasts with the smooth planation surface of the Mio-Pliocene (Brault et al., 2004). The development of the Pleistocene topography of the Armorican massif is explained by

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vertical movements due to the N160 compression generated by the Alpine collision (Bonnet et al., 1998; Müller et al., 1992; Nicolas et al., 1990). This differential tectonic uplift has been estimated from geomorphological studies (Bonnet et al., 2000, 1998; Lague et al., 2000). Incision is measured using a digital elevation model analysis. A distributed field of incised quantities is produced for each individual drainage basin (Jost, 2005). Moreover, relief induced by fluvial incision partly reflects continental movements and can be used to estimate uplift. Indeed, considering that the drainage networks adjust to sea level, the induced incision has the same magnitude in different basins. Thus the difference of incision is used to estimate the relative amounts of differential uplift between basins. The estimation of differential uplift for each basin has been grouped by zone due to the regional organization. A differential tectonic uplift has been observed for the western part compared to the eastern part, along the Quessoy/Nort-sur-Erdre fault (Brault et al., 2004; Jost, 2005).

From these previous work, to reconstruct the paleotopography of the whole area, we first used a digital elevation model (DEM, 100m resolution) given by the Institut Géographique National (IGN) to generate several surfaces providing an estimation of the Pleistocene topography prior to erosion and valley incision (Bonnet et al., 2000, 1998). Within a rectangular 20 km large sliding window, either the highest point or the Q90 quantile was recorded. This process allows for the creation of a surface above the present-time topography connecting the highest points and filling the valleys. The use of the Q90 quantile provides a smoother result than the maximum which may be too influenced by isolated peaks. The generated surface does not include the unknown amount of erosion of peaks and crests and thus should be considered as a minimum. Then, the uplift (Jost, 2005) was subtracted from the preprocessed topography. Last the paleocoastlines were reconstructed for the three transgressions by comparing the reconstructed topography with the expected sea level derived from Hardenbol et al., (1998). The Messinian and Reuverian transgressions had covered a large part of the Armorican massif, except the highest elevations; the last Gelasian/Calabrian transgression had flooded areas along the current coastline, and the lowest inland parts (Figure 14).

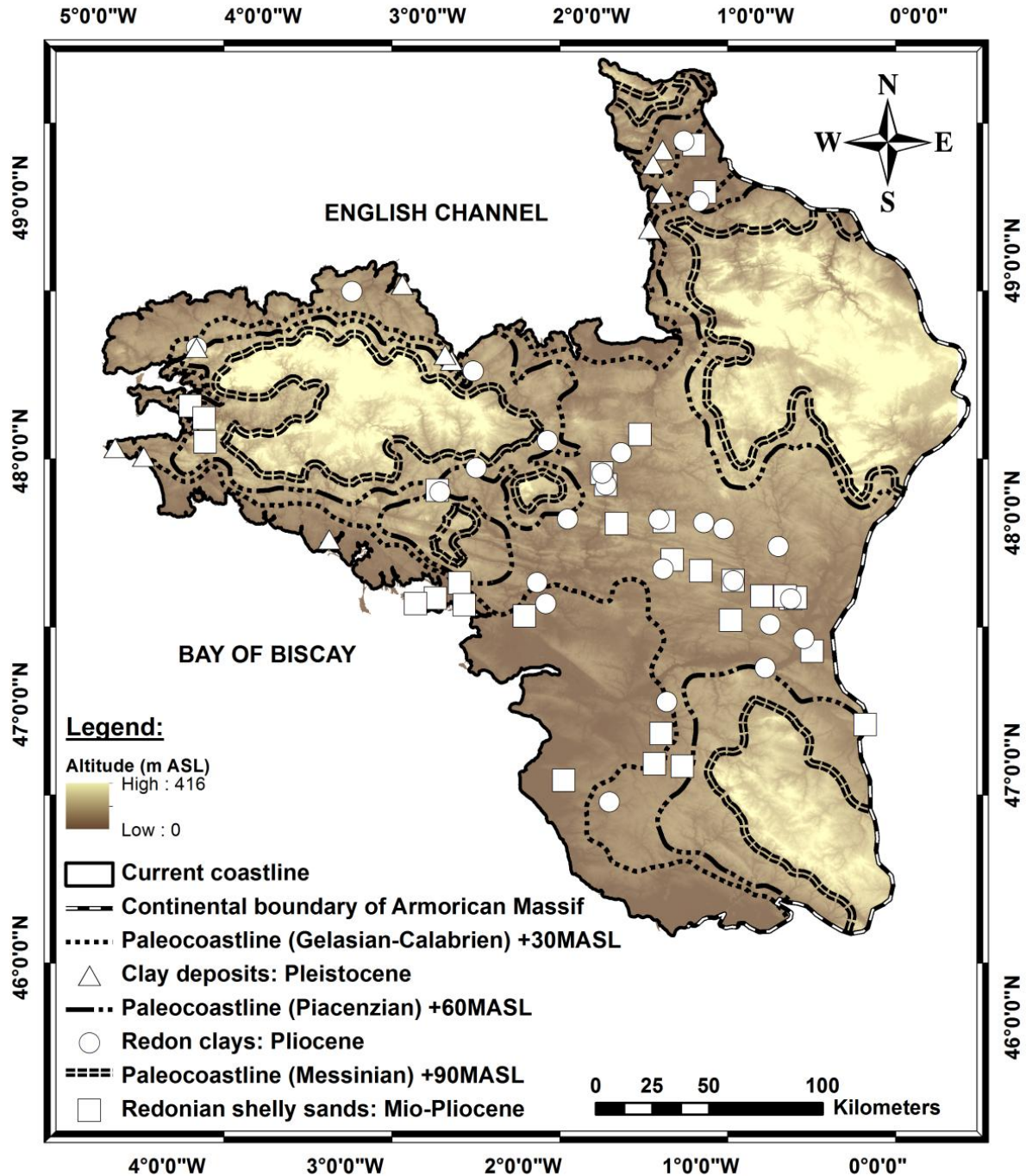


Figure 14. Localization of paleocoastlines on the current topography for past marine transgressions since the Mio-Pliocene time and characteristic sediments associated for each flooding period.

6. RESULTS

6.1. Origin of chlorides

Within the framework of this study, 12 sites (monitoring sites in Figure 12) presenting moderately saline fluids (relative to the high chloride concentration) were subjected to further geochemical and isotopic analysis (only Br concentrations are presented here). Salinities ranging from 60 to 1400mg/l were recovered at depths ranging from 41m to 200m, except for the Cinergy drilling project where water was collected at a depth below 450m in fractured schist, with a chloride concentration of 1240mg/l.

The chloride to bromide relationships is presented in Figure 15 for the 12 sites investigated geochemically. The chloride concentration is linearly correlated with bromide along the rainfall-seawater mixing line (Figure 15). This strongly suggests that the saline fluids in the Armorican basement are of marine origin and correspond to paleoseawater diluted by meteoric waters (Bottomley et al., 1994; Casanova et al., 2001; Frappe et al., 1984; Fritz, 1997; Gascoyne and Kamineni, 1994; Nordstrom et al., 1989).

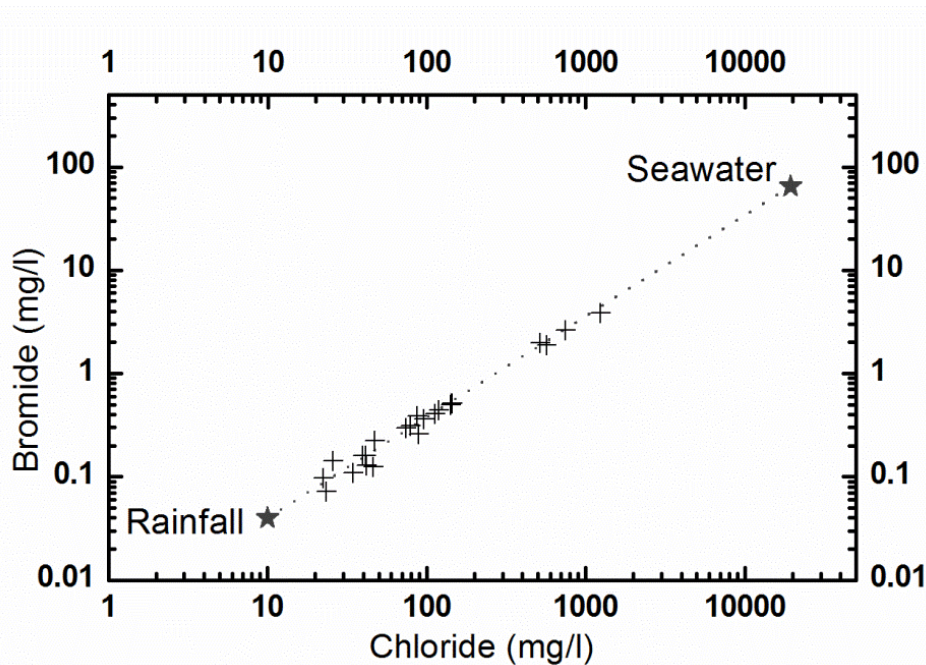


Figure 15. Br versus Cl concentrations of groundwater in the 12 sites investigated.

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Chloride has three potential sources: (1) (paleo)seawater, (2) rainfall potentially concentrated by evapotranspiration processes, and (3) anthropogenic sources (agricultural fertilizers such as KCl, pig slurry and cattle manure). If we exclude a 1-km band across the coastline, the meteoric waters in Brittany have a maximum mean chloride concentration of 10-18mg.l⁻¹ with a relatively low variation range (Ayraud et al., 2008; Martin et al., 2004). Evapotranspiration in Brittany represents about 50-60% of the total precipitation, and cannot provide chloride concentrations in recharge water higher than 40 mg.l⁻¹ (Ayraud et al., 2008). Agricultural pollutions may increase the concentrations to 50mg.l⁻¹ in the most intensively farmed areas (Martin et al., 2004). Thus these two processes cannot explain the very high concentrations observed. Furthermore a negative correlation between nitrate and altitude has been observed. Above 100m, the nitrate concentration is almost constant and below 100m the nitrate concentration decreases with depth. This negative correlation between nitrate and altitude clearly as well as gas groundwater dating (Ayraud et al., 2008) indicates that below this depth the anthropogenic influence decreases. These results underline the fact that the anthropogenic sources are more related to shallow aquifers, contrary to deep saline fluids. Many other sources of chloride exist (as evaporitic deposits of geological formation...). But the relationships between chloride and bromide will be completely different for evaporated brines or evaporite leaching.

Thus, chloride concentrations are clearly out of the potential range of modern waters even including anthropogenic sources. The correlation between bromide and chloride (Figure 15) supports a marine origin for these groundwater (Bottomley et al., 1994; Carpenter, 1978; Freeman, 2007). The simplest mechanism which explains the high chloride concentration observed is a marine component related to the last transgressions.

6.2. Spatial and vertical distributions of chloride

The chloride distribution presented in Figure 16 shows the altitudinal limits of the last three major transgressions between the Mio-Pliocene and the Pleistocene (Messinian ~5.3 Myr, Piacenzian ~2.7 Myr and Gelasian-Calabrian ~1.8 Myr). These limits correspond to 90, 60 and 30m, respectively. This map shows relatively high chloride concentrations (60 to 1400mg/L) around the current coastline and in a few large domains whose current topography is clearly above sea level. Only three zones present chloride concentrations below 40mg/L. These zones correspond to the three domains of higher elevation above 200m and to the area above the higher Messinian paleocoastline altitude of +90m asl. Figure 17 shows the vertical distribution of chloride concentrations according to the altitude of the base of the well. Above 100m, the chloride concentration is almost constant, increasing only slightly with depth, the values ranging from 10 to 40mg/l. Below 100m, some drastic changes in the chloride-depth trends are observed. The main one is apparent at approximately 70m, with chloride concentrations up to 200 mg.l⁻¹ observed below this depth. The chloride concentrations below 70m increase strongly with depth (approximately 0.5mg.l⁻¹.m⁻¹ for the average and more than 1mg.l⁻¹.m⁻¹ for the 90th percentile). This trend contrasts with the almost constant chloride concentrations in the areas not affected by past marine transgressions.

The distribution of chloride concentrations is in good agreement with the paleocoastlines (Figure 16). The increase of chloride concentration with depth (Figure 17) also supports this correlation. The vertical distribution of chloride concentrations likely indicates that, during each of the last transgressions, seawater was introduced by gravity-driven flow at great depth in the basement, and has only partially been flushed by meteoric waters.

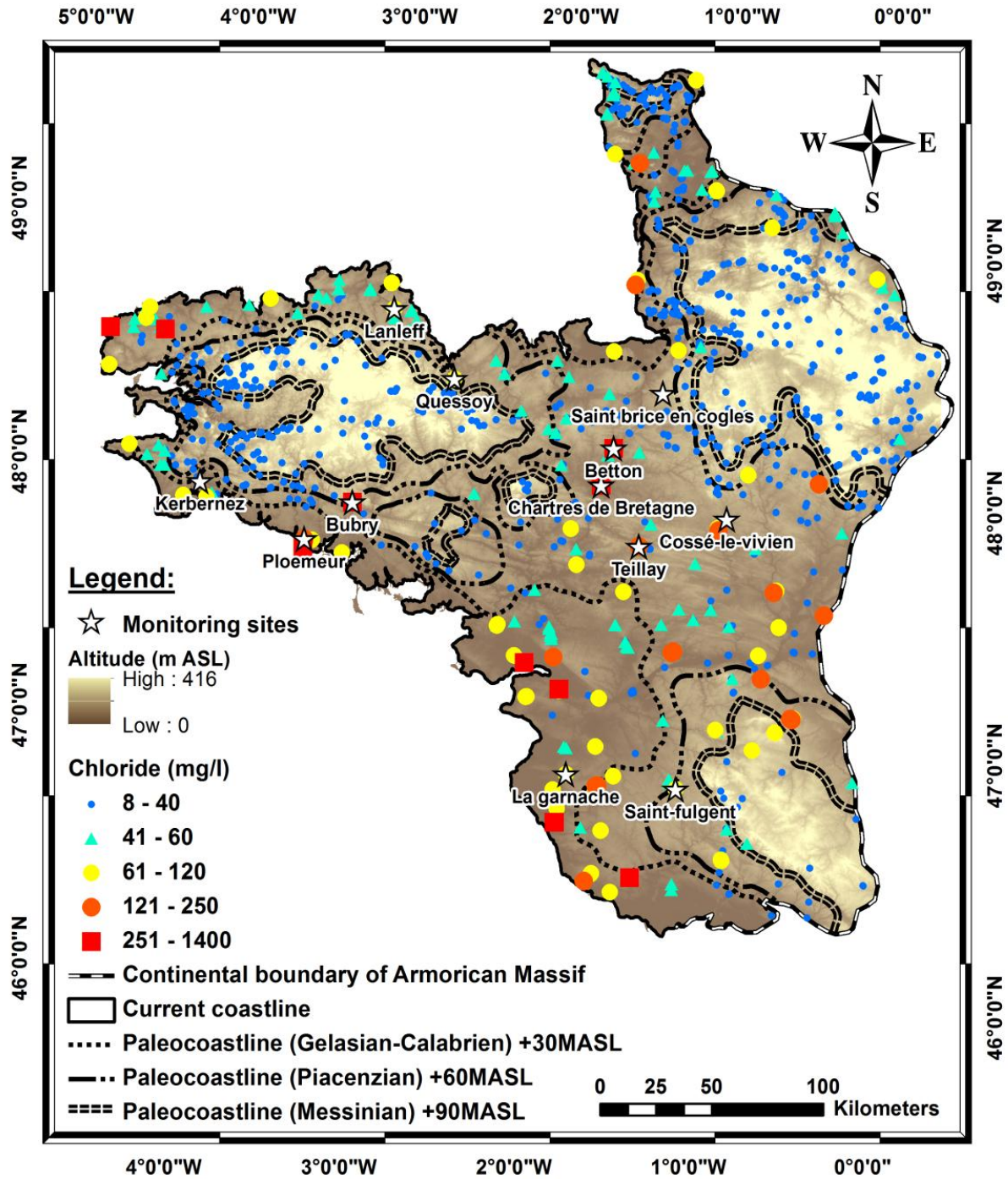


Figure 16. Distribution of paleocoastlines on the current topography for the three transgressions.

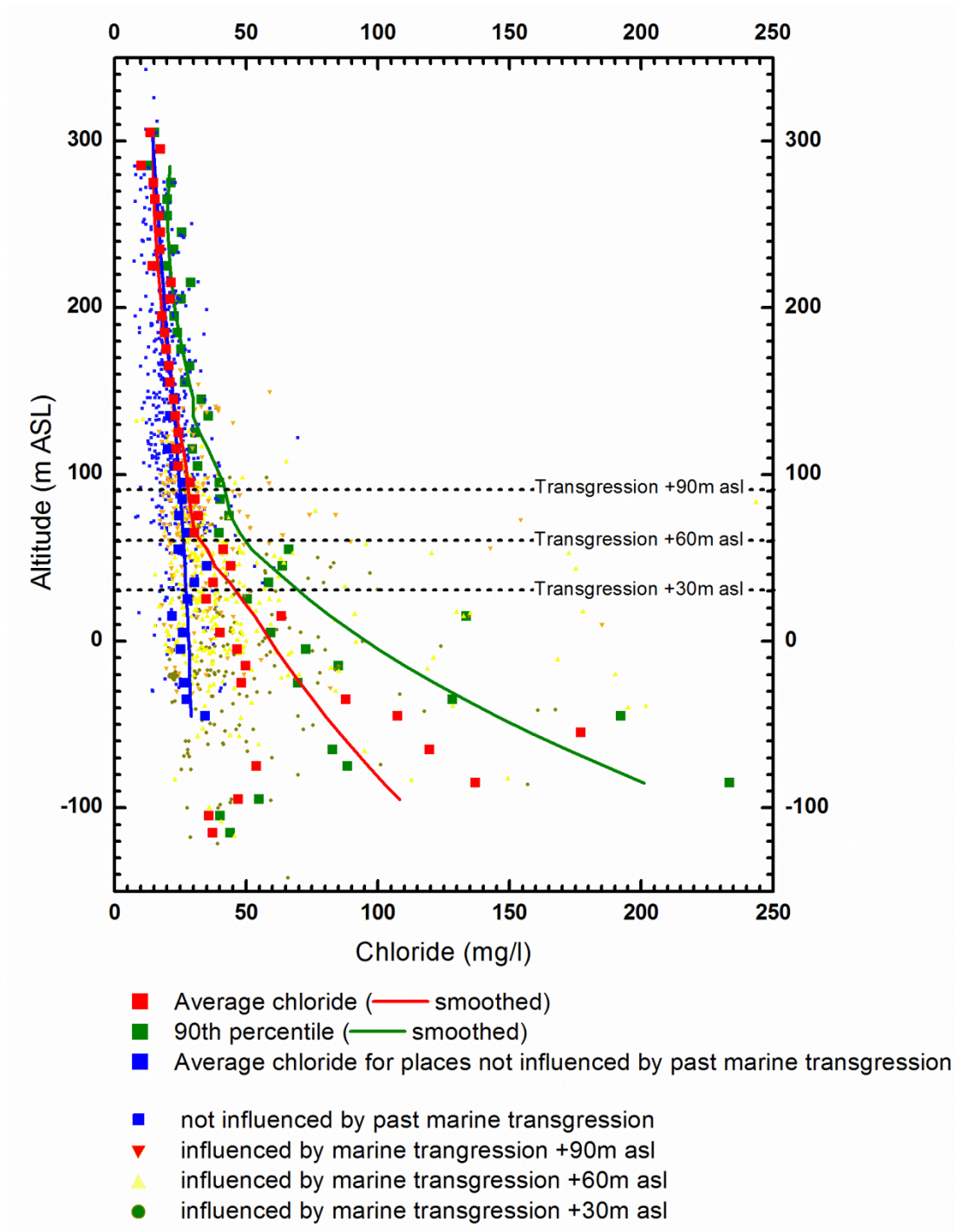


Figure 17. Chloride concentrations versus altitude of well base.

6.3. The chloride releasing rate

The three paleocoastlines of past marine transgressions can be used to define four spatially distinct domains in the Armorican massif: three of them correspond to places where either the Messinian, Reuverian and Gelasian/Calabrian transgressions is the last marine event that covered them; the fourth is the domain that was not flooded Figure 16. The average chloride concentration for each area has been calculated and attributed to the age of the latest marine transgression. Figure 18 shows that the average chloride concentration for each area is inversely related to the time elapsed since the last corresponding marine transgression.

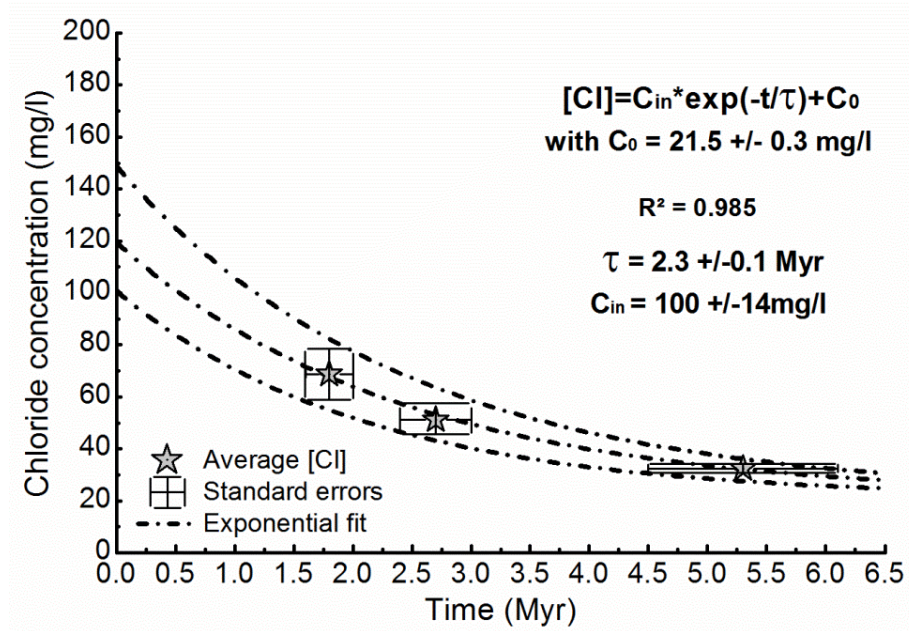


Figure 18. Average chloride concentration for each transgression zone versus the elapsed time since the transgression.

This relationship shown in Figure 18 can be fitted by a simple exponential function of the general form:

$$c = c_{in} \exp - \frac{t_e}{\tau} + c_o, \quad (1)$$

where c is the current chloride concentration, t_e is the age of the last transgression event, τ a characteristic time scale, c_o a background concentration, and c_{in} an additional concentration injected at the time t .

Equation (1) is the solution of the differential equation:

$$\frac{c}{t} = -\frac{(c-c_o)}{\tau} + c_{in}\delta(t_e), \quad (2)$$

$\delta(t)$, the delta function, simulates quasi-instantaneous injection of chlorides at transgression times. The background chloride concentration c_o can be measured in places not influenced by past marine transgression; it is estimated to $21.5 \pm 0.3 \text{ mg.l}^{-1}$. The regression fit leads to a time constant τ of $2.3 \pm 0.1 \text{ Myr}$, and an injected concentration of $100 \pm 14 \text{ mg.l}^{-1}$. The range errors for τ and c_{in} are calculated from the two regression fits generated from the errors on the dating methods used for each characteristic sediments and from the standard deviations of the current chloride concentration of each area. The envelope provided by the two regression fits shows the sensitivity of the parameters according to the standard errors. This residence-time order of magnitude is consistent with the residence time of several million years suggested by (Thury et al., 1994) for deep water as well as several studies supporting long residence times required by "fossil" seawater (Stober and Bucher, 1999). It can be noticed that these studies of fluids containing a paleoseawater end-member have chloride concentrations with an order of magnitude similar to those presented here.

7. DISCUSSION

7.1. General trend of increasing salinities with depth

The main results of this paper present clear evidences on the origin and the age of saline fluids in the continental crust at the scale of the Armorican massif. We report the vertical distribution of chloride concentration which presents a concentration increase with depth at the Armorican massif scale. Such a trend has already been observed in several places throughout the world in continental crust.

Highly saline brines have been found in various geological environments in the Canadian Shield (Frape et al., 1984; Fritz and Frape, 1982; Gascoyne and Kamineni, 1994), in the crystalline basement of Europe (Black Forest in Germany, Soultz-sous-forêt in France) (Aquilina et al., 1997; Pauwels et al., 1993; Stober and Bucher, 1999), in the Fennoscandian shield (Fritz, 1997; Nordstrom et al., 1989) and in the England Carnmenellis Granite (Edmunds et al., 1985). Saline fluids sampled in these sites report a wide range of chloride concentrations. At the surface, chloride concentration ranges

from around ten to hundred mg/L, at much greater depth around 500m concentration values are between one hundred to thousands mg/L and below 1km depth concentrations range from hundreds to tens of thousands mg/L.

For all these sites chloride concentration have been analyzed by intervals of depth where the average chloride concentration is calculated in order to define the concentration gradient until approximately 1km of depth (Figure 19). The Armorican data are also plotted on this graph (Figure 19). All the studies show a general increase of salinity with depth. In the first three hundred meters a high gradient of salinity is observed and between 350 and about 700m, another gradient is observed. Below this depth, a stabilization seems to be observed although there are relatively few data (Figure 19). However the main difference between all studies relies in chloride concentrations of groundwater samples at shallow depths. In fact, chloride concentration can vary by one order of magnitude, with concentrations around ten to hundred mg/L, as for the Armorican basement and the Canadian Shields, respectively (Figure 19).

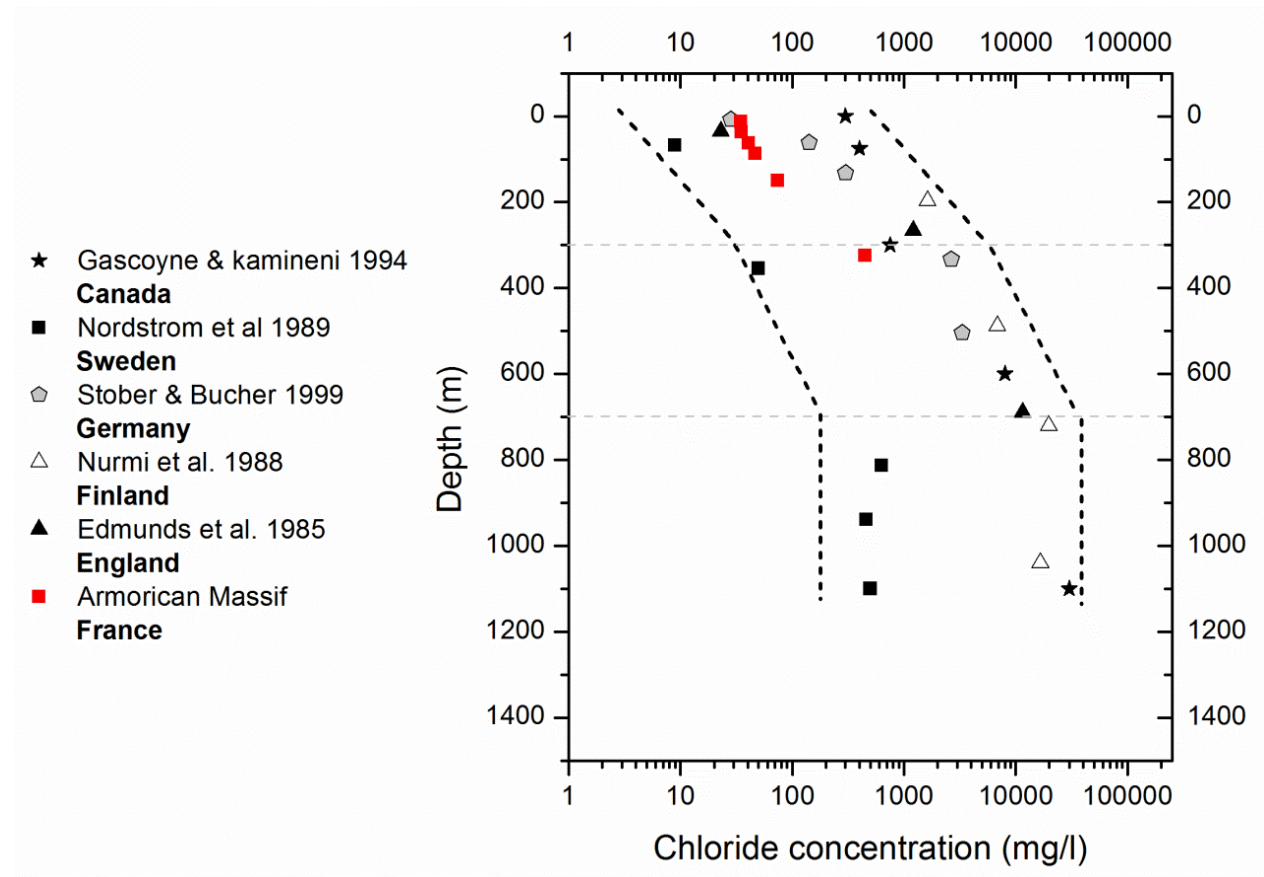


Figure 19. Chloride concentration (mg/l) versus depth (m) recorded in continental basement around the world compared with the data of the current study.

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Previous studies (Aquilina et al., 1997; Edmunds et al., 1985; Frape et al., 1984; Fritz and Frape, 1982; Fritz, 1997; Gascoyne and Kamineni, 1994; Nordstrom et al., 1989; Pauwels et al., 1993; Stober and Bucher, 1999) have been realized in the framework of nuclear repository, geothermal or scientific programs with the aim to investigate the deep continental crust at depths ranging from several hundred meters to several kilometers. In this study we made a synthesis of a large number of wells in a much shallower part of the crystalline basement (41 to 200m). Despite the different purposes, we report similarities, specifically the distribution of chloride with depth which suggests common hydrogeological mechanisms. The salinity gradient from surface to greater depth and the smoothing of the evolution below a certain depth suggest two hydrodynamic behaviors. Such information can be useful to develop conceptual models of basement aquifers functioning. In addition, in this study we provide a time frame constraint of these processes in order to contribute to a better understanding.

7.2. Chloride releasing rate

Figure 18 shows that the relationship between concentration and transgression age can be fitted by a decreasing exponential function, whose parameters give three different characteristics: the background concentration c_o which can be independently evaluated in places that have not been flooded by marine transgressions, c_{in} the additional concentration injected in the system during a transgression, and τ the “dilution” time scale. Note that even if the choice of the exponential function is rather arbitrary considering the small number of constraints, both c_{in} , the curve extrapolation for an event of age 0, and τ , the time to achieve a significant decrease, are rather well determined. However, it is also important to note that situation where three areas have been submerged successively by marine transgressions is exceptional. Even if, the three field-base data points makes for a weak fit, the contribution provides on the saline fluids residence time is clearly better than all previous studies and consequently constitutes a strong constraint. According to the fitting values, the average chloride concentration likely increased by about 100mg.l⁻¹ after each transgression. This is a rather low value compared to the seawater concentration of ~19 g.l⁻¹, which indicates that the original seawater has replaced only part of the freshwater initially present. During transgression, mechanism drives the displacement of former fresh groundwater by seawater, but this process enables the saline fluid circulation at great depth in fractured

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aquifers through the most permeable structures. Nevertheless, all scales of porosity containing former fresh water are not directly influenced. The presence of saline fluid in all scales of porosity requires the diffusion of saline fluid on long term. Thus, the former fresh water already presents has a great contribution on the injected concentration value and has a strong tendency to decrease the value. Then, the C_{in} value estimated in our study is related to the entire volume of the aquifer which tends to reduce the value. And, note that this value corresponds to the increase of the average chloride concentration in the sampling zone that is mostly below 100 m of the current topography. If there is a systematic increase of chloride concentration with depth, as observed in the upper section, the average will increase as well. A deeper sampling could then give higher c_{in} values. The fit also indicates that chloride flushing from the aquifers is a process requiring approximately 2.3 Myr to return to initial conditions.

The leaching process time constrains the long-term hydrodynamics of Armorican aquifers but incorporates many environmental conditions specific to the Armorican massif. Indeed, the efficiency of the leaching process is influenced by numerous physical parameters and paleohydrologic conditions. One of the most important parameter is the topographic gradient controlling gravitational flow and consequently fluid migration. However, meteoric fluid migration into deeper parts of aquifer depends on the hydrostratigraphic configuration of the region. The intensity of topography driven flows is mainly controlled by geographic and climatic conditions. Changes in topographic elevation due to tectonic and erosion processes also impact the behavior of flow systems. In crystalline rock aquifers, as the Armorican basement, tectonic structures (faults and fractures) play a major role through the formation of preferential pathways. Moreover, sea level fluctuations can greatly influence topography driven fluid flow. In the case of an important sea level modification, meteoric fluids may circulate deeper and induce mixing. Thus, the characteristic residence time estimated in our study includes specific physical parameters relating to the Armorican aquifers (geometry of the geological formations, geomorphological features...) and all the changes caused by paleohydrologic conditions since the seawater introduction (tectonic, erosion, sea-level fluctuations, climate changes...). While taking account of the specified points and despite uncertainties on paleocoastlines and the exponential function arbitrarily chosen, the essential point is the clear evidence of past marine transgression during the last millions years and thus leaching processes being active during a similar duration. We use

these characteristic time of hydrodynamic processes to constraint the conceptualization of basement aquifers functioning in the two next sections.

7.3. Conceptualization of Armorican aquifers paleohydrogeology

The Armorican massif presents clear evidences of a marine origin of the saline component in the fluids at the massif scale. During marine transgression, seawater was introduced into the basement by density-driven flow. This mechanism induced the displacement of former fresh groundwater by seawater. Then due to diffusion process taking place on million year (according to the time since transgression) the diffusion length scale (few hundred meters) leads to a perfect mixing between marine waters and freshwaters already present in the whole rock porosity. Third, following transgression, the marine signature contained in the system is flushed out by groundwater circulations fed by meteoric waters. The preservation of marine signature throughout the Armorican basement and the clear increase with depth support the seawater introduction by gravity-driven flow at depth in the basement followed by a flushing phase with limited downward fresh groundwater circulation loops. The flushing process leads to the displacement of marine waters in the upper part of Armorican aquifers. The limited depth of groundwater circulations is explained by the dense hydrological network, the high sea-level and the low topography relief within the Armorican massif. Moreover, the chloride releasing rate provides a time frame for the flushing process which has been estimated in the order (of magnitude) of a few million years. Such information can be used to develop conceptual models of the paleo-functioning of Armorican aquifers. The objective of this section is thus to propose conceptual models that may account for the chloride increase with depth on one hand and for the chloride concentration increase fit with time presented above.

Such information can be used to develop conceptual models of the paleo-functioning of Armorican aquifers. The objective of this section is thus to propose conceptual models that may account for the chloride increase with depth on one hand and for the chloride concentration increase fit with time presented above.

The crystalline basement of the Armorican massif presents aquifers with a high transmissivity associated to fault zones (Roques et al., 2014). Considering the current topography of the Armorican massif including three domains of higher elevation above

200m, the groundwater flow system is driven gravitationally which leads to both local and regional circulation loops (Toth, 1963). Considering the modern situation and constraints provided by the distribution of chloride discussed previously, two main possibilities may explain the distribution of saline fluids in Armorican aquifers.

7.3.1. First conceptual model: Perfectly mixed aquifer

First, the crystalline rock aquifers are considered as a perfectly mixed reactor where the time constant of millions of years is also the residence time of chlorides in the system. The chloride contained in the system is slowly flushed out by freshwater under the influence of topography-driven groundwater flow (Toth, 1963). This model requires that chlorides are located in a compartment of the groundwater system whose recharge is far lower than the current recharge rate (around 200 mm.yr⁻¹) at the top boundary of the system (see Appendix A). This does not necessarily means that this conceptual model is not consistent with data. On the contrary, it seems consistent with the flow partitioning between local and regional flow systems initially presented (Toth, 1963) and precised more recently (Cardenas, 2007; Goderniaux et al., 2013). The characteristics of the partitioning between shallow aquifers and deep aquifers have been studied using the topography of Brittany and assuming a constant permeability with depth (Goderniaux et al., 2013). This study shows four interesting results about the partitioning between shallow (short circulations related to first-order basins) and deep (more regional circulations) aquifers: i) the “deep” compartment can be quite shallow depending on local topography characteristics; ii) the residence time of particles is well fitted by an exponential function, which means that the concentration flushing should behave similarly; iii) the partitioning depends on the total recharge; (iv) the recharge flow in the deep aquifer is 2-5 times smaller than the total recharge. These recharge values are not small enough to explain the 2.3 Myr residence time observed for chlorides, which could mean either that the first model is inconsistent with data, or that the assumption of Goderniaux et al., (2013) is unrealistic. A permeability decrease with depth, may observed in the continental crust with a likely factor of 100 within the first kilometer (Ingebritsen and Manning, 1999; Saar and Manga, 2004; Stober and Bucher, 2007). Such decrease could account for a much smaller recharge of the deep groundwater system and thereby a flushing process less efficient. Through modeling is ongoing to further test this hypothesis.

7.3.2. Second conceptual model: Deep reservoir

In the second case, the presumption is that chlorides could be localized in a deep reservoir i.e. in the deeper part of the aquifer. Then chlorides would be leached out from the aquifer system after diffusion from the deep reservoir towards the active part of the crystalline rock aquifers where leaching is efficient. Calculations for this conceptual model (see Appendix A), lead to a “salt” layer thickness of 40m that does not seem unrealistic as regards the chloride evolution with depth observed. However this conceptual model would predict very high concentrations at depth that are not yet observed in the Armorican basement in the absence of very deep drillings. But, this could be in agreement with highly saline fluids sampled in various geological environments at great depths worldwide (Figure 19). As in the crystalline basement of the Black Forest where Stober and Bucher (1999) suggest that the deep salt water discharges is related to the upwelling of deep water which may bring up chloride from a deep reservoir. However, it can certainly be affirmed that the salt contained in the deepest part is not solid due to the chloride to bromide relationships which excludes this possibility.

The next step of this study will be to test these two functioning hypothesis with numerical modeling in order to provide more realistic models. The objective is to provide some constraints on the saline fluid circulation at the regional scale as well as to understand if the heterogeneity and hydrodynamic conditions may induce a wide range of leaching rates which could explain the large range of chloride concentrations observed.

8. CONCLUSION

Chloride concentrations in groundwater were investigated in the Armorican basement at shallow depth (from +350 to -150masl). The high concentrations observed in some places (from 60 to 1400mg/L) are shown to originate from marine transgressions. The distributions of high chloride concentrations are in good agreement with both the altitudinal and vertical limits and succession of the three major transgressions between the Mio-Pliocene and Pleistocene ages. During each of the last transgressions, seawater was introduced by gravity-driven flow at great depth in the basement. This seawater end-member has only partially been flushed by meteoric waters (over 1 to 2 million years) between successive marine transgressions, and since the last one. Considering the paleocoastlines, three zones have been affected by the transgressions. The average chloride concentration calculated for each area is inversely related to the time elapsed since last marine transgression. This relationship between concentration and transgression age can be fitted by a decreasing exponential function which indicates that chloride flushing from the aquifers is a process requiring approximately 2 Myr. The fate of chloride is conceptualized using two main models: a dilution or leaching of a deep reservoir. Both models provide information on the history of the saline fluids in fractured aquifers. The first model requires a drastic partitioning between shallow and deep aquifers. In the second model, chlorides are supposed to be released by diffusion from a deep reservoir, leached by fresh groundwater flow and bringing up to the shallower aquifer. The model supposes a deep reservoir, which has not yet been identified in the Armorican massif, but could exist. This study thus provides important constraints on the seawater circulation in hardrock aquifers at a continental-scale area.

Appendix A

The objective of this section is to develop simple calculations that aim at figuring out the meaning of the data and fit presented before. We first assume that the average of chloride concentrations measured is related to the average chloride concentration in the aquifer used in the model. Because of the increase of chloride concentration with depth and because of the limited sampling depth, it is unreasonable to pretend that the average of the measurements equals the chloride average concentration in the entire aquifer. Thus, we just guess that the chloride average in the entire aquifer evolves with a similar time scale to the measured averages, which occur for instance if the depth dependency function is similar in different places. Regardless of the model, the general evolution of the chloride concentration is given by a mass balance equation of the type:

$$\frac{d(V\bar{c})}{dt} = -Q_c + Q_{in} \quad (3)$$

where V is the system volume, \bar{c} the average chloride concentration in the system, Q_c the total flow at the system discharge boundary, and Q_{in} the total flow at the system recharge boundary. The “system” can be the entire aquifer or any part of it. Q_{in} represents a potential chloride inflow from the recharge boundary. If we assume a background concentration c_o , Q_{in} can be written as:

$$Q_{in} = \iint_{S_r} r(s) \cdot c_o ds = c_o \cdot A_r \cdot \bar{r} \quad (4)$$

where S_r is the system recharge boundary, $r(s)$ the recharge at any point s of the boundary, A_r the recharge boundary surface, and \bar{r} the average recharge flow.

We then define two end-member models that may explain the chloride evolution in basement aquifers.

A1 The perfectly mixed aquifer

In the first model, the chloride contained in the deeper part of the aquifer is slowly flushed out from the system along the discharge at a rate Q_c such as:

$$Q_c = \iint_{S_d} q(s) \cdot c(s) \cdot ds \quad (5)$$

where S_d is the system discharge boundary, $q(s)$ the flow discharge at any point s of the boundary, and $c(s)$ the chloride concentration. In a perfectly-mixed reactor, the total concentration at the boundary is equal to the concentrations average. Even if this approximation is certainly unlikely in highly heterogeneous aquifers, we assume that the Q_c integral can be estimated from the averages of concentrations and discharge Q_c defined as:

$$Q_c = O \cdot A_d \cdot \bar{q} \cdot \bar{c} \quad (6)$$

where \bar{q} and \bar{c} are the discharge and concentration averages, respectively, and A_d is the total surface of the system discharge boundary. $O(1)$ is a constant equal to 1 in the perfectly-mixed approximation, and whose order of magnitude is about 1 if the approximation is not rigorously exact. The water mass-balance requires that the total discharge equals the total recharge: $A_d \bar{q} = A_r \bar{r}$. If we assume that the aquifer system is a rectangular box fed from above with a recharge r , using a thickness $h = \frac{V}{A_r}$, equation (3) writes as a first-order kinetic equation :

$$\frac{\bar{c}}{t} = \frac{(c_o - O(1) \cdot \bar{c})}{\tau} \quad (7)$$

with the time constant τ equal to $\frac{h}{r}$.

If we consider that r is of the same order as it is now in Brittany (about 200mm.yr⁻¹), given the time constant τ of 2.3 Myr, we predict an unrealistic aquifer depth h of about 500km. Although the actual depth is not known, it is likely less than a few km and more likely about 1 km. This means that the dilution model is valid only if the recharge is at least 2 to 3 orders of magnitude less than the current rainfall.

A2 The deep reservoir diffusion model

In the second model, we assume that chlorides are leached out from the aquifer system by diffusion from a deep layer of thickness h' without any advection. Q_c is a bit different from equation (7) because diffusion occurs i) on all the system boundaries, and ii) at a rate equal to $q(s) = D \frac{\partial c(s)}{\partial n}$, where n is the direction perpendicular to the system boundary. It is not easy to estimate simply $q(s)$. We assume that the concentration gradient establishes in a length scale of the order of the smallest aquifer dimension h' , which is a reasonable assumption if the aquifer thickness is small compared to its horizontal extent Q_c is defined as :

$$Q_c = O \cdot A \cdot D \cdot \frac{(\bar{c} - c_o)}{h} \quad (8)$$

with O' an “order-of-magnitude” constant similar to O . The influx Q_{in} is now nil and the general equation writes as:

$$\frac{d\bar{c}}{dt} = \frac{(c_o - \bar{c})}{\tau'} \quad (9)$$

$$\text{with } \tau' = O'^{-1} \cdot \frac{h'^2}{D}.$$

The molecular diffusion D_m of chloride in water is on the order of $2 \cdot 10^{-9} \text{ m}^2 \cdot \text{s}^{-1}$ (Li and Gregory, 1974; Wang et al., 1953). Considering a porosity of 1%, the diffusion D of chloride is on the order of $2 \cdot 10^{-11} \text{ m}^2 \cdot \text{s}^{-1}$. A time constant of 2.3 Myr is thus consistent with a salt layer thickness of about 40 m.

Chapitre 2. Impacts des variations climatiques passées. Implication dans la compréhension du système hydrogéologique.

2.3. Impact of climate changes during the last 5 million years on groundwaters in basement aquifers

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Keywords: Climate Change, groundwater resources, saline fluids, aquifers, fluid transfer,

paleohydrology

Section: Physical sciences / Earth, Atmosphere and Planetary sciences

Significance statements: Chemical and isotopic investigation of groundwaters in aquifers in the Armorican basement (western France) at depth of 80 to 400m, allowed the characterization of ubiquitous paleofluid signatures. These signatures demonstrate that climate changes during the last 5 Ma induced introduction of saline marine fluids related to seawater level elevation during warm periods, and glacial melt water during the last glacial period. These fluids circulate in these basement aquifers along fractures through density-driven circulation. Then they occupy the whole rock porosity through diffusion processes. Conversely, present-day aquifers only function in a depth-limited section and have only slightly affected the paleofluid signature. This study thus emphasizes the vulnerability of groundwater resources to climate changes.

ABSTRACT

Climate change is thought to have major effects on groundwater resources. There is however a limited knowledge of the impacts of past climate changes such as warm or glacial periods on groundwater although marine or glacial fluids may have circulated in basements during these periods.

Thorough geochemical investigations of groundwaters at shallow depth (80 - 400m) in the Armorican basement (western France) revealed three major phases of evolution : (1) Mio-Pliocene transgressions led to marine water introduction in the whole rock porosity through density and then diffusion processes, (2) intensive recharge after the glacial maximum down to depths of several hundred meters, (3) a present-day regime of groundwater circulation limited to shallow depth.

This work identifies important constraints regarding the mechanisms responsible for both marine and glacial fluid migrations and their preservation within a basement. It defines the first clear time scales of these processes and thus provides a unique case for understanding the effects of climate changes on hydrogeology. It reveals that glacial water is supplied in significant amounts to deep aquifers even in permafrosted zones. It also emphasizes the vulnerability of modern groundwater hydrosystems to climate change as groundwater circulation is restricted to shallow depths.

Introduction

The last glacial period resulted in drastic modifications of the hydrogeological regime (McIntosh et al., 2011; Person et al., 2007). Sedimentary basins show evidence of glacial recharge (Edmunds and Smedley, 2000; Klump et al., 2008; McIntosh et al., 2002, 2011; McIntosh and Walter, 2006; McMahon et al., 2004; Stute and Forster, 1995). The case for fractured aquifers in basements is less clear. Modeling of the hydrogeological regime in the Fennoscandian and Canadian shield during the last 10,000 yrs, has also indicated potential phases of glacial groundwater circulation in the basement (Lemieux et al., 2008a, 2008c). However, whether recharge of glacial water is continuous or not during the last glacial maximum is not known. Furthermore, relationships of recharge with permafrosted areas remain unclear as well as recharge mechanisms (Castro et al., 2007; Stotler et al., 2010; Zongyu et al., 2003).

In recent decades, basement saline fluids have been sampled at great depths (0.5 - 5 km) in the context of nuclear-waste disposal investigations, and other general research programs. There is now evidence that such fluids are relatively ubiquitous in basements worldwide (Bucher and Stober, 2010; Frape et al., 2003). However their origin remains difficult to decipher due to intensive water-rock interactions and the extremely long residence-times (Stotler et al., 2010). In several cases, analyses with geochemical tracers have suggested that the solutes are of marine origin (Aquilina and Dreuz, 2011; Aquilina et al., 2000; Beaucaire et al., 1999; Bottomley et al., 1999, 1994; Louvat et al., 1999). There is however no clear evidence of a potential marine component in hard rock aquifers. As an example, some of the brines in the Canadian shield are suggested to be of Pleistocene cryogenic origin (Herut et al., 1990; Starinsky and Katz, 2003) but are also suggested to be of marine origin, which would require a Devonian age (Bottomley et al., 2003, 1994; Katz and Starinsky, 2003).

The mechanisms that could explain how and when marine fluids could be introduced into the basement remain poorly understood (Greene et al., 2008; Stotler et al., 2012). Furthermore, no knowledge of the mechanisms involved in the preservation of marine or glacial fluids and the time length of such preservation has been available until now.

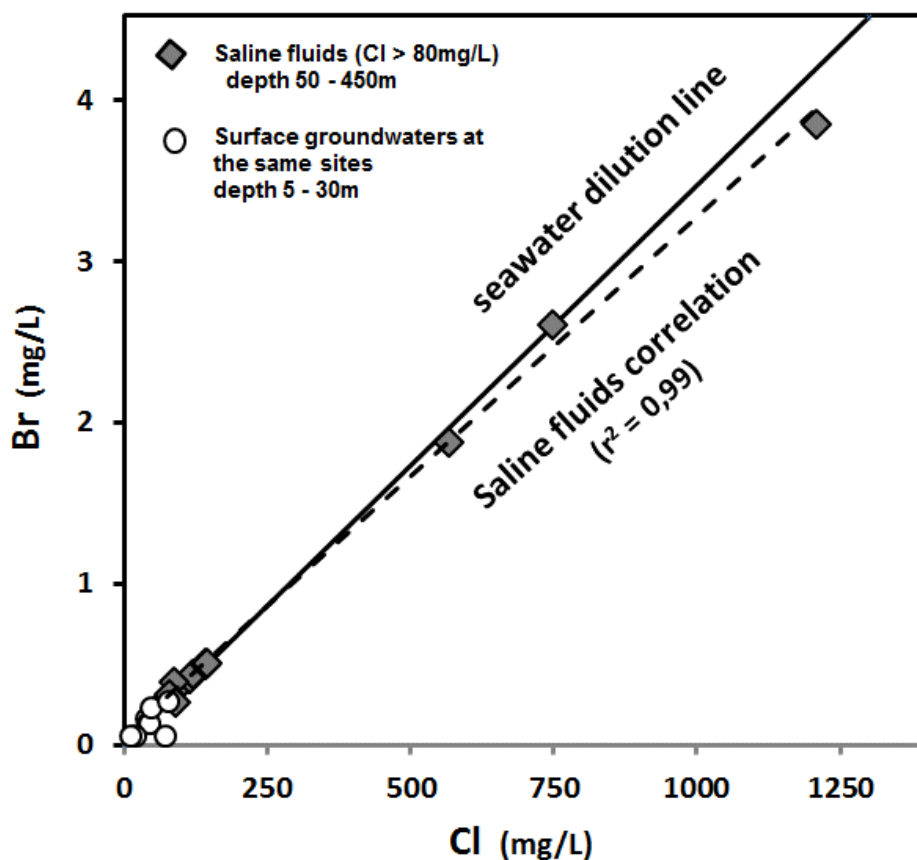
Basement rocks also constitute major groundwater resources which, until now, have been little exploited. However, there is now growing pressure for more intensive use of these resources. In recent decades, anthropogenic pressure has resulted in major drawdown and chemical evolutions (Custodio, 2002; Galloway et al., 2004) that accentuates the modifications induced by modern global warming as temperature increase should limit groundwater recharge in a large part of the earth surface before the end of this century (Green et al., 2011; Scibek and Allen, 2006). Deciphering the effects of climate change on paleo-hydrogeological regimes in basements is therefore of great importance. More precisely, the effects and ages of marine transgression on fluid circulation in basements need to be reliably identified, and the velocity of solute leaching together with the effects and duration of glacial fluid circulation quantified.

1. Geological and hydrogeological setting

The Armorican basement covers an area of 68,500 km² in western France. It consists of Precambrian and Paleozoic rocks from the Cadomian and Hercynian geodynamic cycles. The altitudes reach 400 m in places but commonly range from 100 to 200 m. Most of the geological formations are fractured and folded granites, gneiss and schists. Cretaceous and Tertiary weathering has produced a weathered layer, several tens of meters thick. From a hydrogeological point of view, this weathered layer constitutes an aquifer which is highly sensitive to diffuse agricultural pollution (Aquilina et al., 2012). Additional aquifers can be found in the fractured rocks below this aquifer where water circulation is related to fault zones (Ayraud et al., 2008).

More than 1,800 wells designed for groundwater exploitation are recorded in the ADES database (<http://www.adeseaufrance.fr/>). This database was used to analyze the spatial distribution of chloride concentrations in wells located further than 1 km from the coast line (Armandine Les Landes et al., 2014).

Twelve sites in the area with very high chloride concentrations (> 80 mg/L) were selected. Groundwater was collected after pumping at depths of 80 to 150 m. One sample was collected from a deep research well ("Cinergy sample") which intersected the basement at 405 m below a local sedimentary basin, down to a depth of 750m. This was the deepest and most saline sample ($\text{Cl} = 1,208$ mg/L). A sub-group of three sites (Bubry, Betton and Cinergy sites) with the most saline samples ($\text{Cl} > 150\text{mg/L}$) is defined. A detailed description of the analytical methods is given in supplementary information (Analytical methods section).



2. Results

2.1. Solute origin

Whereas the Cl/Br mass-ratio range in the shallow and modern groundwaters (< 30 m depth) was very large (100 to 600) the Cl/Br mass-ratio of the saline fluids (Cl > 80 mg/L) collected in the Armorican basement was 283 +/- 16 (Figure 20). This ratio is very close to that of seawater (288). The linear Br to Cl relationship ($R^2 = 0.957$) closely matches a dilution line ranging from the deepest, most saline fluid (Cinergy sample) to the most dilute shallow ones. This relationship is equal to the dilution line between the most dilute shallow samples and a modern seawater end-member. The boron isotope value for the more saline fluid (Cinergy) was + 40.7‰, which is very similar to the isotopic composition of boron in seawater (+ 39‰). The boron isotope ratios in all the other saline fluids were close to this signature except in one sample where the ratios were lower, possibly as a result of interaction with clay-minerals.

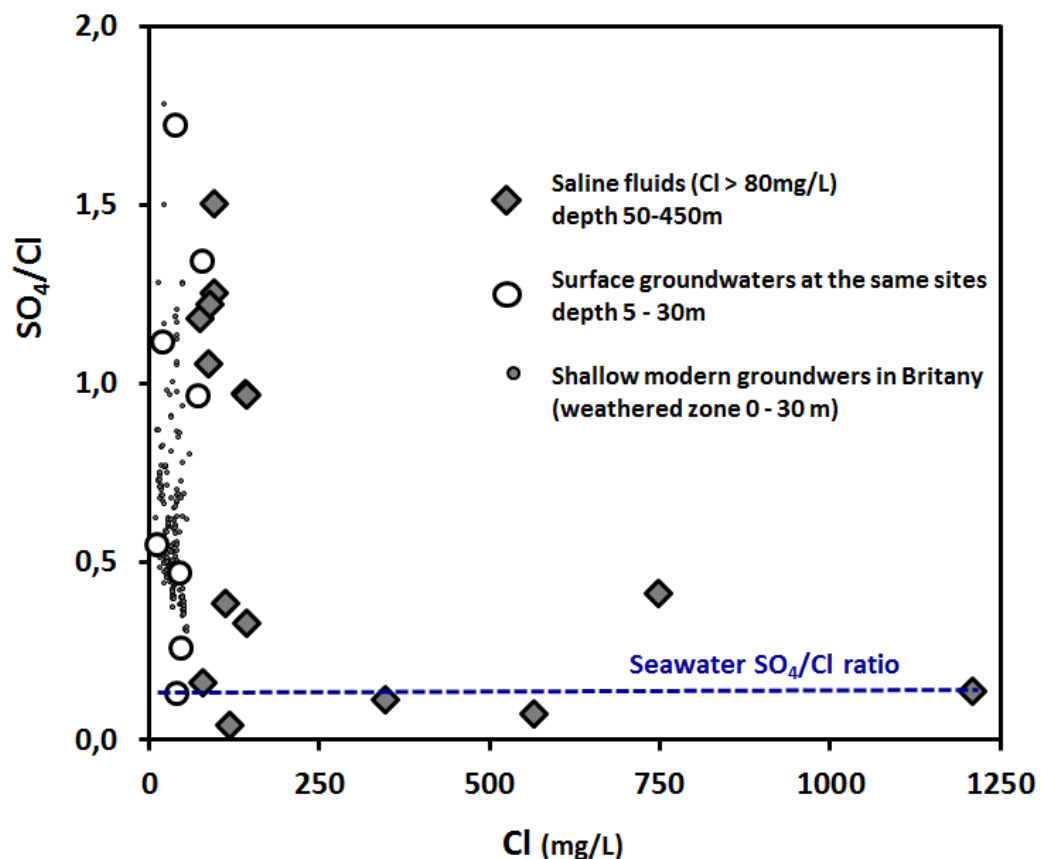


Figure 21. Sulphate to Chloride ratios (modern groundwater, data from the Agrhys observatory).

Chapitre 2. Impacts des variations climatiques passées. Implication dans la compréhension du système hydrogéologique.

Modern and shallow groundwaters present a wide range of SO_4/Cl mass-ratios (0.3 to 1.8, Figure 21) generally far above the marine mass-ratio (0.14) and the SO_4/Cl mass-ratio in local precipitation (0.44 ± 0.3). Oxidation of sulphide minerals such as pyrite, ubiquitous in igneous or metamorphic rocks enhances the sulphate concentrations in the shallow groundwater. Furthermore, nitrate constitutes a major electron acceptor in modern agricultural catchments, whereas the oxidation mechanisms in groundwaters that are not anthropogenically influenced are much more limited (Böhlke and Denver, 1995; Tarits et al., 2006). The sulphate concentrations in all the collected saline samples were high with SO_4/Cl mass-ratios ranging from 0.04 to 0.4 with a mean of 0.2 ± 0.14 when Cl concentrations overpasses 150mg/L, close to the ratio found in seawater (0.22).

Different domains are apparent in the $\delta^{18}\text{O}_{\text{SO}_4}$ vs $\delta^{34}\text{S}_{\text{SO}_4}$ plot (Figure 22). The $\delta^{34}\text{S}_{\text{SO}_4}$ values for shallow groundwaters in Brittany, particularly nitrate-contaminated groundwater, are within the range of +8 to +16‰. Groundwaters influenced by sulphide dissolution present lower isotopic ratios (down to -10 ‰) as the isotopic ratios for sulphide $\delta^{34}\text{S}_{\text{SO}_4}$ are usually negative (Pauwels et al., 2010). $\delta^{34}\text{S}$ of the most saline fluids ($\text{Cl} > 150\text{mg/l}$), ranges from +23 to +25‰. The other saline fluids can be plotted along a mixing line between two end-members: (1) a ^{34}S depleted end-member containing sulphate originating from sulphide oxidations (denitrified fluids in Figure 22) and (2) the most saline fluid. Both $\delta^{34}\text{S}_{\text{SO}_4}$ (+22‰ to +25‰) and a $\delta^{18}\text{O}_{\text{SO}_4}$ (+13 to +16‰) of the most saline fluid exceed that of present day seawater (around +21‰ and +9.5 ‰, respectively). Subsequent 0.8‰ and 6‰ decline of both $\delta^{34}\text{S}_{\text{SO}_4}$ and $\delta^{18}\text{O}_{\text{SO}_4}$ marine sulphate have taken place over the last 3Myrs. Therefore, the dataset indicates values for saline fluids slightly above those reported for 2 to 5 Myr seawater. However the slight difference can be explained either by a diagenetic reduction of sulphates within the first few centimeters of the sediment column or the occurrence of transitional environments such as lagoons and marshes.

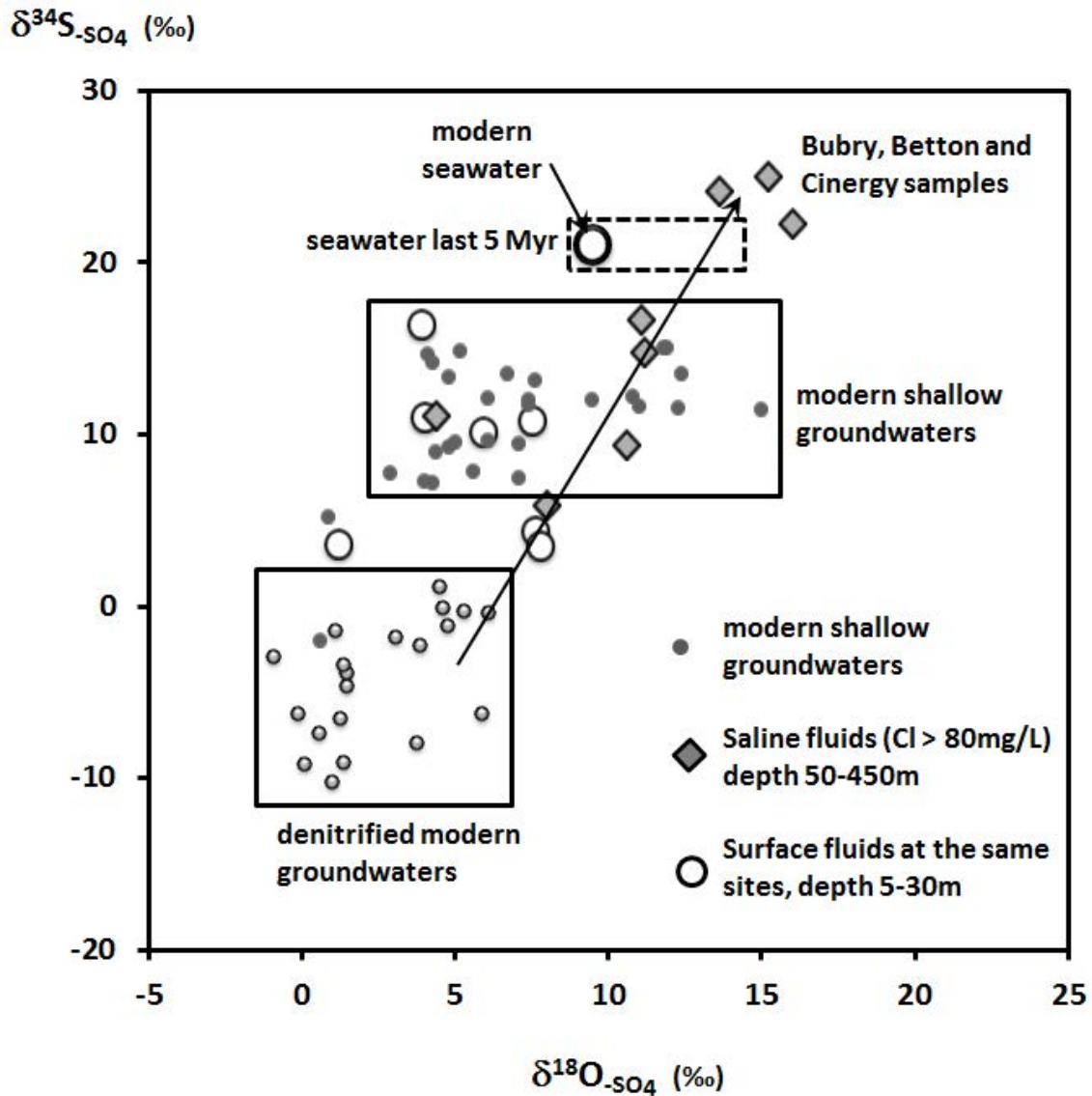


Figure 22. Sulphate isotopic ratio (modern groundwater, data from Pauwels et al., 2010).

2.2. Chloride vertical distribution

The chloride concentrations of all the Brittany wells in the ADES database have been plotted against altitude (asl) in Figure 23. A 10 m average is presented (red curve). A clear increase with depth is observed, especially below 100 m asl. The mean chloride concentrations in the highest 250 m (i.e. above 100m asl), vary between 15 and 30 mg/L. Such concentrations are indicative of meteoric and anthropogenic sources (Armandine Les Landes et al., 2014). The sharp chloride increase observed below 100 m requires a third source.

A more detailed investigation revealed that the chloride concentrations precisely reflect the last three transgression events (Armandine Les Landes et al., 2014). This result strongly supports a marine origin for the water solutes below 100 m depth, whatever the site.

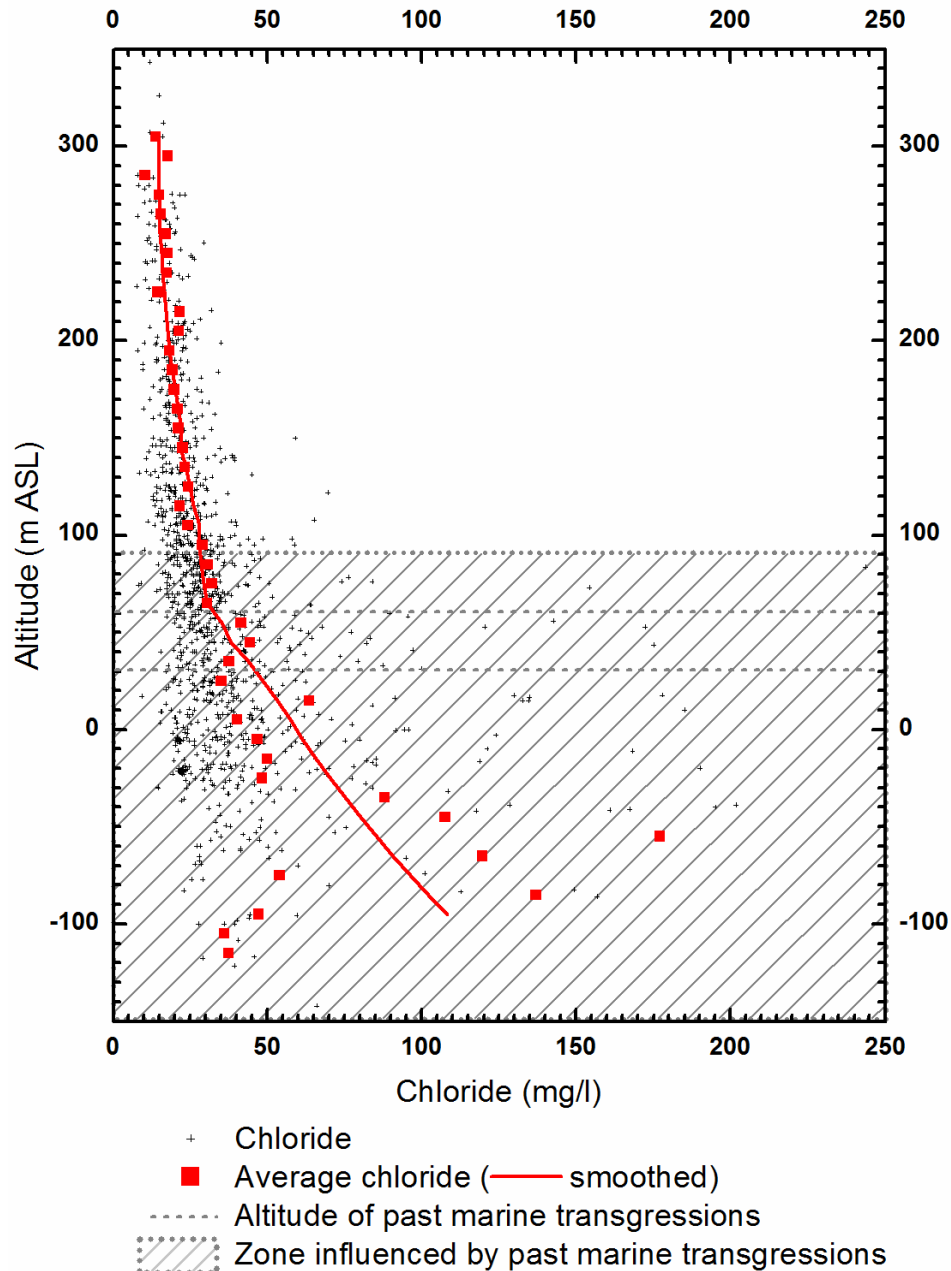


Figure 23. Chloride vertical distribution

2.3. Recharge temperature

The stable isotopes in the most saline fluids can all be plotted along the local meteoric water line (LMWL) and represent meteoric continental waters (Supplementary information, stable isotopes section). Only the composition of the deepest and most saline Cinergy-sample differs from the LMWL. When possible, the most saline fluids were compared with shallow ground-waters at the same location. A systematic shift towards more negative values was observed along the LMWL. Such a shift indicates that the recharge conditions were colder than modern ones. This shift is correlated to both the chloride concentration and the recharge temperature (Supplementary information, stable isotopes and noble gas section) which favors the temperature interpretation to other potential explanations (altitude, continental effect...).

Recharge temperatures were deduced, using an inverse model, from the Ar and Ne concentrations (Aeschbach-Hertig et al., 1999) (supplementary information, stable isotopes and noble gas section). The recharge temperatures for the collected fluids ranged from 5 to 10 °C, except for the deepest and most saline Cinergy sample which presented a recharge temperature close to 0 °C and also extremely high excess air. The present-day recharge temperature in Brittany is about 12 °C and the noble gas measurements in shallow groundwater agree with such temperatures (Ayraud et al., 2008). It is thus clearly apparent from the recharge temperatures that the recharge conditions were colder than today. This is particularly true for the deepest and most saline Cinergy sample which would have required glacial or immediately post-glacial recharge conditions.

A clear correlation was observed between recharge temperature and chloride concentrations (Figure 24). The three most saline samples display a linear correlation between surface shallow groundwater and the deepest and most saline Cinergy sample.

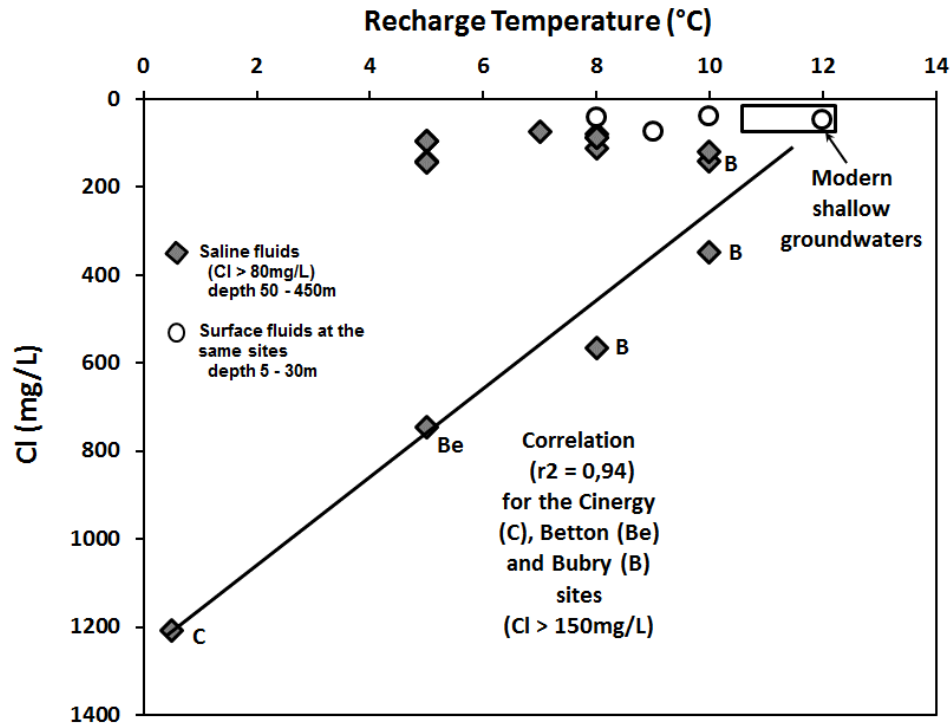


Figure 24. Recharge temperature deduced from noble gases vs chloride concentration

2.4. Groundwater dating

CFC groundwater dating shows that modern groundwater presents residence times close to 20yrs in the weathered part of the aquifers (0-30m) and 20 to 50yrs and more in the deeper part (Ayraud et al., 2008). All the saline fluids present CFC concentrations close to the detection limits which is interpreted as “old” fluids (> 50yrs) mixed with a limited amount of modern surface waters. On the basis of the nitrate and CFC content, the amount of modern surface fluids contained in the samples ranges from less than 1% to 15% (supplementary information, Table S1). ^{36}Cl and ^{14}C analyses were carried out within 3 samples including the two most saline ones (Cinergy sample Cl = 1,208mg/L; Betton sample Cl = 700mg/L). A detailed interpretation of these data is provided in supplementary information (Section Groundwater dating, Figure S1 and Table S3 and Table S4). ^{36}Cl in these samples is more likely at the equilibrium with the neutron production of the Proterozoic formations which indicates residence times of at least 10^6yr . Conversely, ^{14}C indicates residence times of 17,100 to 17,600yrs for the two most saline fluids. These data are interpreted as representing two different events as presented below. The chemical basis for the scenario is also given in supplementary information (mixings section).

3. Discussion

3.1. A two-phase evolution

As already suggested by several studies of old, saline fluids, the solute and water may have different origins and represent different phases of fluid circulation corresponding to geologic or global climatic changes. They will therefore be discussed separately. The agreement between chloride vertical distribution and the altitudes of the last transgressions, from 5.3 to 2 Myrs old, supports a marine origin of the solutes in groundwater below 100m asl. High Cl concentrations, requiring a marine contribution, are observed throughout the Armorican basement except in the zones preserved from past transgressions at altitudes higher than 90m asl where the Cl concentrations remain below 50 mg/L. This correlation is also supported by the geochemical relationships, Br/Cl and SO₄/Cl mass-ratios, as well as the B and S isotopic ratios. The marine signature of the solutes from saline fluids in the basement thus represents the generalized influence of transgressions prior to the glacial period at the scale of the Armorican basement. ³⁶Cl data that suggest secular equilibrium are in good agreement with the transgression timing.

It was concluded in several earlier studies of saline fluids worldwide that the solutes were of marine origin (Aquilina and Dreuzy, 2011; Aquilina et al., 2000; Beaucaire et al., 1999; Bottomley et al., 1999, 1994; Louvat et al., 1999). However it was difficult to confirm this due to intensive water-rock interactions. In contrast, this signature is clearly apparent in the Armorican aquifers for various solutes (Cl, SO₄, Br, B...). This may be related to the very short time since the last transgression.

On the contrary, the noble gases and water stable isotopic ratios clearly demonstrate that the water in these saline fluids is of meteoric origin and that recharge occurred during glacial or immediately post-glacial conditions. Glacial water strongly diluted the older seawater already contained in the basement rocks. We interpret this contradiction as reflecting a two-stage evolution namely: (1) Introduction of seawater-derived fluids into the basement 5.3 to 2 Myrs ago, which constitute the source of the solutes collected in most of the wells below 100m asl in the Armorican basement. (2) Recharge of cold meteoric water. Glacial melt water introduction follows the end of the last glacial maximum 19,000 yrs ago at the onset of deglaciation of the permafrost in northwestern

Europe 18 to 17,000 yrs ago (Jiráková et al., 2011). Although it might be possible that glacial recharge has occurred several times during the past, following the various glaciation cycles, ^{14}C ages mainly agree with a limited period at the onset of the last deglaciation.

A hydrogeological model based on these observations is depicted in Figure 25 and presents this two-phased circulation and the very different modern hydrogeological regime. These 3 situations are discussed below.

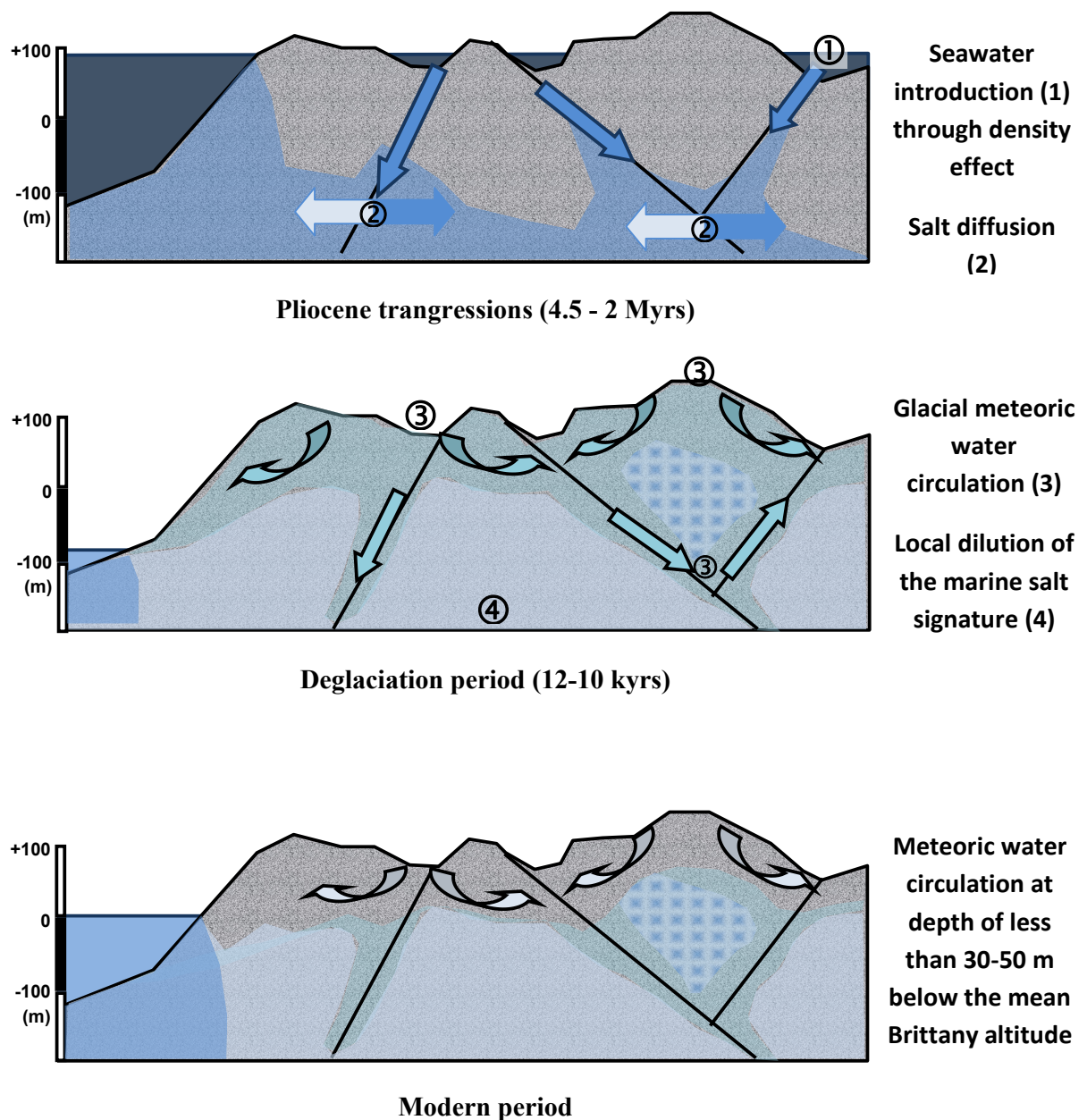


Figure 25. Hydrogeological model derived from the geochemical data

3.2. Seawater introduction into the basement

The attribution of a marine signature to several deep saline samples worldwide indicates that the mechanisms responsible for seawater introduction into basements occur relatively frequently. The introduction of saline fluid into basements requires drivers. These may be upward fluxes related to basin brine expulsion. A typical example of such mechanisms is the fluid migration responsible for MVT-type metal deposits. However, such fluid circulation seems to be mainly restricted to the basin/basement interface (Aquilina and Dreuz, 2011; Boiron et al., 2002). As transgression occurs, the simplest mechanism of seawater introduction into the basement is by gravity- and density-driven displacement of former fresh groundwater by seawater. Such a process may account for saline fluid circulation at relatively great depth in the most permeable fractures. However, this mechanism cannot explain the relatively ubiquitous occurrence of saline fluids throughout the Armorican basement. Furthermore, the most saline fluids were collected in the Cinergy well at a depth of 405-750m below a small Mesozoic sedimentary basin, away from the border faults of this graben. Thus the process was thorough and efficient, allowing the saline fluid to penetrate all the scales of porosity and not only the major structures.

Molecular diffusion can be estimated from the diffusion of water molecules in water D_m , which is about $10^{-9} \text{ m}^2 \cdot \text{s}^{-1}$ for typical groundwater temperatures. The diffusion length scale – i.e. the length of the mixed zone – is about $\sqrt{\theta D_m t}$, where t is the time since recharge and θ is the porosity ranging from 0.01 to 0.1, leading to a mixing length of about 20 to 60 m for 1 Myr, and from 2 to 6 m for 10kyr. It would seem from these estimates that the marine chlorides are probably well mixed at the scale of the investigation, i.e. a few hundred meters. If seawater had been introduced into the basement at higher time-scales than the million years diffusion scale, it should have diffused throughout the hard rock porosity. Even the immobile water in the microporosity would contain marine solutes. This does not mean that the concentration is constant throughout the aquifer since (1) the flow is likely rapidly renewed in the upper part of the geological section, and (2) an increase in concentration, due to the density effect, should still be observed as the time required for complete homogeneous diffusion of the entire domain exceeds several Myrs. In particular, a depth gradient

should be observed as the density-driven flow should result in a greater influence at depth.

3.3. Glacial recharge

In contrast, glacial meteoric recharge presents the characteristics of a relatively rapid and violent process. First, the deepest and most saline Cinergy sample contains extremely high excess air. Such excesses are thought to be characteristic of rapid recharge of glacial melt (Corcho Alvarado et al., 2011; Klump et al., 2008), which would agree with the low recharge temperatures. This trapping of excess air implies a rapid and catastrophic recharge event. Second, the preservation of these glacial signatures in all the fluids investigated indicates a generalized process which has not been erased by further mixing with modern groundwater. It is interpreted as a rapid and short recharge event which occurred between 18 and 17,000yrs ago after the glacial maximum. This period corresponds to the first retreat of northwestern glaciers and could also correspond to the breakthrough of the permafrost in Brittany (Jiráková et al., 2011). Resumption of recharge processes have also been observed in sedimentary aquifers in northwestern Europe at a similar period (Corcho Alvarado et al., 2011). During this period, the seawater level was about 80m below its present level, which would have allowed deeper penetration than the modern groundwater regime.

Hydrodynamic dispersion, which is the second process that might have diluted the original glacial or marine signal, is classically modeled by a diffusion equation. The diffusion coefficient in this equation is the product of the average flow velocity v by the hydrodynamic dispersivity length A . The dispersion length is thus equal to \sqrt{Ax} , where x is the distance of flow since recharge. The distance from recharge zones is reasonably between 100m and 1000m for most groundwater samples. Hydrodynamic dispersivity values of between 10^{-2} and 10^3 m have been reported in field scale studies but the reliability of values greater than 1 is poor (Gelhar et al., 1992). The resulting estimated dispersion length is between 1 and 30 m. In contrast with molecular diffusion, the mixing length scale is only dependent on the flow distance and not on the time since the recharge event. This back-of-the-envelope calculation explains why we can have in the same place relatively high chloride concentrations, signatures of marine transgressions during warmer climates, as well as a glaciation recharge signature.

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In contrast to the marine signature, the signature of the last glaciations has not been diluted at a distance of more than two meters. Although glacial water may exchange solutes with the microporosity at a local scale, it did not have time to equilibrate through diffusion with the surrounding rocks. Major fractures allow water circulation and microporosity acts as a reservoir for solutes. If water circulation is slow, the water in the fracture may equilibrate through diffusive processes with solutes from the microporosity immediately around the fracture without modifying the water signature (Aquilina et al., 2011; Waber and Smellie, 2008). However, the glacial signature will remain much more heterogeneously distributed than the much older marine signature.

The correlation between recharge temperature and chloride concentrations also supports this interpretation even though these parameters are related to two distinct processes (marine transgression and glacial melting). It shows that the last major circulation events were triggered by conditions extremely different to present conditions. Density-gravity-driven circulation is required to inject marine solutes at depth and the glacial recharge may be related to a much lower seawater level and much deeper piezometric levels in the continent. Such lowering of the water table could have allowed efficient recharge as the groundwater could not be discharged into the river network which was too far from the watershed surface. The most saline and deepest samples present the highest salinity and the coldest recharge temperature, as the original zones of marine signature, and also reflect the most rapid and active circulation of post-glacial water with high excess air and low recharge temperatures.

Recent modeling of groundwater infiltration during the glacial cycle indicated that major infiltration occurred below the glacier during the ice sheet progression whilst it remained limited in the permafrost areas (Lemieux et al., 2008b). Exfiltration also seemed to dominate during the deglaciation period. Geochemical observations may however indicate more complex processes in the periglacial areas (Stotler et al., 2012). The Armorican basement is considered to have been a discontinuous periglacial permafrosted area during the last glacial cycle (Jiráková et al., 2011). The evidence of ubiquitous glacial water about 17,000yrs ago rather indicates a continuous permafrost and a massive breakthrough at the onset of the glaciers retreat before the generalized deglaciation.

3.4. Modern recharge

Although the salinities are not high, in comparison to the original seawater, the homogeneous preservation of salinity throughout the Armorican basement and the clear increase with depth indicates the limited downward circulation of modern groundwater. Circulation loops are mainly constrained by potential discharge zones. The depth of groundwater penetration is limited both by the surface hydrological network and the high sea level (as compared to during the glacial period). The non-existence of high elevation recharge zones within the Armorican basement, which could have flushed the deep saline fluids, has allowed the signatures of both the marine transgression and the glacial circulation events to be preserved.

Conclusion

Investigation of a large database of chloride concentrations in the groundwater of hard rock aquifers in the Armorican basement in western France, revealed higher chloride concentrations below 100m asl than could be expected from meteoric and anthropogenic sources. Twelve sites presenting chloride concentrations higher than 80mg/L were investigated for isotopic, geochemical and dissolved gases analyses.

These data are interpreted as reflecting: (1) Introduction of marine seawater during the last transgressions 5.3 to 2 Myrs ago through gravity- and density-driven infiltration. This introduction was followed by diffusion of the marine solutes throughout the entire hard rock domain at the scale of several hundred meters, and exchanges with the rock microporosity. (2) Rapid recharge of meteoric water at the onset of deglaciation. Glacial water circulated in the main active fluid circulation zones. This water may have equilibrated with the surrounding rocks during the last 17,000 yrs thus allowing a single sample to contain both marine solutes and meteoric glacial solutes. (3) Following these two phases, modern groundwater circulation seems to be depth-limited as no dilution of these signatures has occurred.

This study indicates that marine transgression leads to a generalized introduction of seawater into hard rock aquifers. Diffusion allows the marine signature to be present throughout the area. It should thus be quite a common component of groundwater in hard rock aquifers. Conversely glacial recharge seems a rapid event, related to a

restricted period related to the breakthrough of the permafrost. Contrary to common belief glacial water may circulate even in permafrosted areas, at least during short time periods. The presence of a glacial signature throughout the Armorican basement indicates that glacial water, probably glacial melt water, has a huge impact on groundwater even if it represents a time-limited event.

As compared to saline fluids in basement elsewhere, the Armorican fluids agree with the overall depth-salinity relationship (Armandine Les Landes et al., 2014). The most striking difference is the relatively shallow depth of these fluids, probably induced by the lack of relief, and the relatively recent age of the transgressions. These two point have allowed both the marine and glacial signatures to be preserved however the scenario described should have a large applicability, at least as test hypothesis, in crystalline rocks worldwide. The Armorican saline fluids thus confirm that fluid signatures in basements may record the various signatures of geologic and climatic events. They can be considered as archives of "crisis" events at the geological scale which are recorded in the basement aquifers. It also emphasizes the role of fault-zones in the fluid transfer at depth. Our results also indicate that the extent of hydrogeological circulation of fresh water has been limited to less than 50 to 100m during the last 17,000 yrs. Thus our findings emphasize the high sensitivity of groundwater resources to global climate changes as well as to anthropogenic pressure. They provide time-constraints for groundwater modeling under various climatic scenarios.

Acknowledgments

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Supplementary information

Analytical methods section

Wells were pumped and physico-chemical parameters (pH, Eh, T°C, Cond.) were monitored after stabilization was reached. Samples were collected in polyethylene bottles rinsed with ultrapure water and acidified for cation analysis. Anions, cations and trace-elements were analysed in the Geosciences Rennes Laboratory using chromatography (anions) and ICP-MS (cations and trace).

Dissolved gases were collected without air contamination in steel bottles using a specific Grundfoss pump. CFC and SF₆ were measured in the Rennes Condée-Eau analytical laboratory through gas-chromatographic analysis with ECD detector. Ne and Ar gases were measured through gas-chromatographic analysis with a catharometric detector. Uncertainty is about 3% for CFC and SF₆ and about 5% for other gases.

Stable isotopes were determined in the BRGM Laboratory through equilibration with $\delta^{18}\text{O}$ and δD uncertainties of ± 0.1 and 0.5‰ . Sulphate isotopic compositions were determined using a Delta S mass spectrometer (Thermo Finnigan). The $\delta^{34}\text{S}$ of sulphates was measured from SO₂ obtained from CdS precipitated after sulphate reduction and $\delta^{18}\text{O}_{\text{-SO}_4}$ was determined from the CO₂ produced by the reaction of BaSO₄ with C at 1050°C. The Canon Diablo Troilite and Vienna-SMOW standards were used for S and O isotopes respectively with uncertainty about $\pm 0.3\text{‰}$.

$^{36}\text{Cl}/^{35}\text{Cl}$ ratio was measured by AMS at the French national ASTER facility. Chemical separation of Cl was conducted based on the protocol of (Conard et al., 1986). To prevent S isobaric interferences, BaSO₄ was precipitated by adding BaNO₃ and was removed by filtration. Next, AgCl was precipitated by adding AgNO₃ and recovered by centrifugation. To improve SO₄ removal, the AgCl precipitate was dissolved in NH₄ before repeating the procedure. AgCl is then precipitated again by adding NH₄OH. A blank solution was prepared with Merck Standard NaCl 99.91%. The measured blanks provided $^{36}\text{Cl}/^{35}\text{Cl}$ ratios that were less than 10^{-15} at/at.

The carbon species (i.e. CO₂, TDIC and carbonates) were converted into CO₂ by direct acidification, and the ^{13}C contents were measured by mass spectrometry (SIRA) at the IDES Laboratory (University of Paris Sud). The ^{13}C content is reported using δ (‰)

notation, as a deviation from the V-PDB (Vienna-Belemnite from the Pee Dee formation, North Carolina, USA). Graphite sources for ^{14}C analyses were prepared from TDIC in the IDES Laboratory, and measured using an accelerator mass spectrometry (UMS LMC14, Gif-sur-Yvette, France). The ^{14}C contents are expressed as a percentage of modern carbon (pmC). Analytical errors, including laboratory errors, are of $\pm 0.2 \text{ ‰}$ vs V-PDB for the $\delta^{13}\text{C}$, and between 0.1 and 0.3 pmC for the A_{14}C .

Mixings section

(1) Influence of modern groundwaters. The saline fluids result from a complex mixing history that is somewhat difficult to decipher. As the saline samples are located far from the sea-coast and present reduced conditions they are thought to represent isolated parts of the aquifers. These samples contain almost no nitrate and CFC concentrations close to or below the detection limit (Labasque et al., 2006). From the CFC and nitrate content a range of mixing of the saline samples by modern surface fluids has been computed assuming deep saline fluids do not contain any anthropogenic tracers. Except for the Ploemeur site which is an intensively pumped site, mixing rates are below 4 % (Table S1). For the three most saline sites (Bubry, Betton and Cinergy), mixing are below 4% for Bubry and below 1.6 and 1.4 % for Betton and Cinergy samples. Such mixings result from extremely slow renewal rates of these fluids or slight contamination during sampling.

(2) Seawater component. In Table S1 are also presented the percentage of seawater necessary to explain the high salinity of the fluids. Percentages of seawater remain low, below 1% for most of the fluids. For the three more saline sites (Bubry, Betton, Cinergy) they are of 0.6 to 2.7 (Bubry), 3.85 (Betton) and 6.25 (Cinergy). Our hypothesis assumes a strong dilution of the paleo-seawater by glacial fluids. The very high content of solutes in seawater as compared to aquifer groundwater explains that the marine signature of Cl, Br, SO_4 and B has been preserved although saline fluids have been strongly diluted (see Figure S1). Even a dilution of seawater by meteoric fresh water degree of 90% only induces a 0.5‰ shift of the mixing $\delta^{34}\text{S}$.

(3) Dilution of the seawater component by glacial fluids. As mentioned above, the saline fluids contain almost no modern aquifer groundwater. The low percentages of seawater in the saline samples have almost no effect on the oxygen and deuterium ratios, which explains that the waters have a meteoric signature for the stable isotopes. However, a part of aquifer groundwater older than 60yrs cannot be distinguished from glacial meteoric water. Only the oxygen-deuterium shift and the recharge temperature presented in the next section indicate the glacial signature. The percentage of glacial water versus non glacial aquifer groundwater cannot be determined. As mentioned in the previous section, a strong dilution effect is consistent with the preservation of the marine signature of various solutes or isotopes, due to the high seawater concentration. For the ^{14}C , on the contrary, the low carbon content of seawater only slightly affects the ^{14}C content of the glacial and/or meteoric fluids (10% of seawater induces a lowering of 2% of the ^{14}C content).

The test of the various mixing hypothesis is consistent with the scenario described in the paper. A residual seawater component of a several percents allows the marine signature of ^{36}Cl , ^{11}B , Cl/Br , ^{34}S to be preserved. A strong dilution by glacial fluids will provide a meteoric signature for the stable O and D of water as well as ^{14}C ages consistent with the glacial dilution period.

Stable isotopes and noble gas

O and D measurements are provided in Table S2. Except the most saline Cinergy sample ($\text{Cl} = 1,208\text{mg/L}$), all the samples plot along the global meteoric water line, defining a potential local meteoric water line. This results demonstrate that the water has a meteoric origin which agrees with the low percentage of seawater contained in the samples (Table S2). However, when compared to the signature of the modern surface aquifer at the same place, the saline samples show a shift towards lower O and D values. To enlighten a potential temperature effect, this difference to the local surface reference has been computed. For the Cinergy sample, the shift is the difference between the fluid collected at the end of drilling (drilling fluid made of local tap water) and the final saline fluid produced after pumping.

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The deuterium shift is plotted against the recharge temperature deduced from noble gases in Figure S2. A good correlation is observed between these two parameters. For oxygen, the correlation is a bit less clear ($R^2=0.57$).

A recharge temperature has been deduced from the Ne and Ar content of the saline fluids. Although less precise than using a larger number of noble gases, the robustness of this computation has been tested. Furthermore, a more thorough analysis of all noble gases was carried out (mass spectrometry, East Anglia Laboratory) for one of the most saline fluid (Betton Pz6 sample, Cl = 747mg/L) and confirmed the noble gas recharge temperature obtained using Ne and Ar analysis.

Table S1. Chemical analyses of the saline fluids from the Armorican basement

Sample	pH	Eh (mV) mesuré	O2 (mg/L)	Cl (mg/L)	Br (mg/L)	B (µg/L)	SO4 (mg/L)	SO4/Cl	Cl/Br	NO3 (mg/L)	DIC (mg/L)	CFC-12 pmol	CFC-11 pmol	CFC-113 pmol	Ne 10 ⁻⁸ mol/L	Ar 10 ⁻⁵ mol/L	10 ⁻³ T°C	% Seawater	% Mixing modern gw
Non saline in the upper aquifers																			
Ploemeur F35 (<50m)	5.90	140	2.4	73.45	0.32	22.30	70.82	0.96	230.00	1.97	21.10	12.06	8.37	0.302	1.53	2.05	9.0	0.38	
Ploemeur F9 (<50m)	6.00	179	6	47.37	0.22	29.90	12.16	0.26	210.79	45.30	3.90	5.40	6.50	0.38	0.98	1.80	9.0	0.24	
Quessoy Captage (10m)	6.10	102	0.12	39.70	0.16	17.6	68.32	1.72	248.13	1.27	11.3	15.95	2.40	0.08	1.20	1.95	10.0	0.21	
Teillais F3 (<50m)	6.64	-54	0.8	41.99	0.13	na	5.36	0.13	323.00	0.8	8.7	1.14	0.22	0.042	1.56	2.1	8.0	0.22	
Betton Pz3 (15m)	5.90	91	6.5	46.23	0.13	11.50	21.60	0.47	369.84	49.61	11.40	2.208	3.819	0.289	0.970	1.510	12.0	0.24	
Saint-Fulgent (50m)	6.96	-3.4	0.91	79.02	0.31	8.80	12.63	0.16	254.40	3.09	29.60	0.39	0.15	0.03	1.57	2.12	8.0	0.41	
Cossé-le-vivien (15m)	6.97	-20	0	88.88	0.26	27.5	108.34	1.22	341.85	5.91	40.4	1.21	0.14	0.03	1.04	1.90	8.0	0.46	
Deep saline fluids																			
Ploemeur F11 (85m)	6.30	143	1.8	74.38	0.30	47.75	87.94	1.18	251.79	1.03	15.20	1.561	0.291	0.0599	1.41	2.12	7.0	0.38	9.3
Ploemeur F38 (80m)	6.50	166	2	87.39	0.39	70.00	92.24	1.06	226.85	7.44	15.60	0.645	0.227	0.0387	1.27	2.01	8.0	0.45	9.6
Bubry F6 (100m)	na	na	na	347.00	1.20	373.00	39.7	0.11	293.00	0.1	17.4	na	0.189	0.047	1.15	1.92	10.0	1.79	1.2
Bubry F5 (100m)	7.5	91.6	1.5	565.44	1.88	410	40.71	0.07	300.77	11.19	21.3	na	na	na	1.04	1.87	8.0	2.92	12
Bubry F3 (100m)	8.31	-49	0.4	119.00	0.44	79	5	0.04	270.45	2.46	17.3	0.333	0.177	0.048	1.1	1.88	10.0	0.61	4.6
Betton Pz6 (90m)	7.20	-34.9	2.7	746.87	2.61	561.00	306.85	0.41	286.16	0.10	30.70	0.03	0.13	0.03	1.58	2.31	5.0	3.86	1.6
Cinergy (400m)	7.69	145.3	3.36	1208.00	3.85	624	166.20	0.14	313.77	0.62	53.5	2.31	0.06	0.09	4.98	3.60	0.5	6.24	1.4
Quessoy Forage (60m)	6.60	89	0	95.93	0.36	86.4	120.28	1.25	266.47	1.15	24.8	3.27	0.14	0.02	1.64	2.29	5.0	0.50	16.8
Teillais F1 (135m)	5.67	-20	1.4	112.80	0.408	47.1	43.36	0.38	276.47	13	3.75	1.33	1.35	0.09	1.76	2.25	8.0	0.58	6.3
Teillais F2 (135m)	5.45	127	4.15	142.40	0.51	53.4	46.4	0.33	279.22	9.2	5.2	1.71	2.03	0.107	1.72	2.15	10.0	0.74	3.6
La Garnache (70m)	6.87	-1	0.15	96.11	na	159.00	144.46	1.50	na	<0,5	58.50	0.49	0.10	0.04	1.25	2.10	5.0	0.50	4.4
Lanleff FE19 (118m)	7.55	146.5	bdl	141.89	0.5	63.8	137.97	0.97	283.78	5.74	33.7	2.02	0.54	0.30	1.26	2.13	5.0	0.73	3.2
Lanleff FE20 (100m)	7.54	160	bdl	143.76	0.51	na	139.1	0.97	281.88	5.64	33.1	1.70	0.46	0.33	1.20	2.07	5.0	0.74	3.4

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Table S2. Isotopic analyses of the saline fluids from the Armorican basement

Sample	$\delta^{34}\text{S}$ (‰) (SO_4)	$\delta^{18}\text{O}$ (‰) (SO_4)	$\delta^2\text{H}$ (‰)) (H_2O)	$\delta^{18}\text{O}$ (‰)) (H_2O)	$\delta^{11}\text{B}$ (‰)) (H_2O)
Non saline in the upper aquifers					
Ploemeur F35 (<50m)	10.8	7.5	-31.4	-5.4	34.3
Ploemeur F9 (<50m)	16.4	3.9			44.5
Betton Pz3 (15m)	11	4.4	-39.5	-6.34	26.5
Saint-Fulgent (50m)	3.5	7.8	-36.6	-5.7	35.3
Cossé-le-vivien (15m)	3.6	1.2	-41.6	-6.3	33.9
Deep saline fluids					
Ploemeur F11 (85m)	5.9	8	-31.2	-5.31	35.5
Ploemeur F38 (80m)	14.8	11.2	-32.4	-5.41	36.6
Bubry F5 (100m)	24.2	13.6	-37.4	-5.9	8.7
Betton Pz6 (90m)	22.3	16	-42.7	-6.82	33.9
Cinergy (400m)	22.3	16	-35.3/-34	-4.7/-4.6	
La Garnache (70m)	16.7	11.1	-34.3	-5.3	16.2
Lanleff FE19 (118m)	11.1	4.4			24.9

Table S3. 36-chlorine measurements

	Total Cl (mmol/L)	^{36}Cl (10^{-15})	$\text{Cl}^{36}/\text{Cl}^{35}$ (10^{-14})	Uncertainty $\text{Cl}^{36}/\text{Cl}^{35}$ (%)
Ploemeur F38	2.507	1.27	5.07	5.02
Betton PZ6	20.986	5.27	2.51	7.17
Cinergy	26.479	6.45	2.44	7.19

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Table S4. 14-carbon measurements and residence time computations

Sample	$\delta^{13}\text{C}$ (‰ vs PDB)	$A^{14}\text{C}$ (pmc)		pH	HCO_3 (mmol/L)	$\delta^{13}\text{C}$ at equilibrium (‰ vs PDB)	Age - Mook model (yr BP)	Mixing with modern surface fluids (%)	Age corrected for mixing (yr BP)
Non or slightly saline fluids in the upper aquifers									
Ploemur F9	-20.83	95.0	± 0.2	5,6	0,68	-21.0	Modern		
Ploemur F11 (85m)	-18.46	77.8	± 0.2	5,4	1,27	-18.2	Modern		
Ploemur F37 (85m)	-19.06	82.1	± 0.2	6,3	1,58	-22.3	1600		
Ploemur MF2	-19.08	84.7	± 0.2	6	1,23	-20.7	Modern		
Saint Brice F3	-18.27	81.71	± 0.3	5,8	1,44	-19.1	Modern		
Most saline fluids									
Ploemur F38 (80m)	-17.64	61.1	± 0.2	6,6	1,78	-22.7	4300	15	6500
Betton P26 (85m)	-15.93	15.6	± 0.1	7,2	2,56	-23.6	16600	1	17100
Cynergi (400m)	-18.31	26.5	± 0.15	10	4,46	-27.1	16000	5	17600

^{14}C has been measured in three saline samples and in non or slightly saline samples at depth ranging from 50 to 85m in two sites. The carbon isotopic composition (^{13}C , ^{14}C) of total dissolved inorganic carbon (TDIC) in groundwater is acquired mainly during transit in the unsaturated zone by exchanges with soil CO_2 and the carbonate matrix (Gillon et al., 2009). For all samples, the calculated $\delta^{13}\text{C}$ of the gaz in equilibrium with the water TDIC do not evidence any equilibration with the carbonate matrix in the saturated zone which provides a good confidence to the age computation. The high pH of the Cinergy sample induces a very low equilibrated $\delta^{13}\text{C}$ but has limited consequences on the age computation. Considering Fontes (1992), the appropriate adjustment model corresponds to the one proposed by Mook (1980) (Fontes, 1992; Mook, 1980). It has been applied to all data (Table S4). A correction has been introduced using the mixing with surface modern groundwater defined from NO_3 and CFC concentrations. The activity of the Ploemur F9 sample (highest ^{14}C activity) has been used as a modern end-member to correct the A_0 activity. No correction has been introduced for the $\delta^{13}\text{C}$ or the pH as the mixing percentages are low. Ages have also been computed using an equilibrating model (Fontes and Garnier, 1979) showing a relatively low uncertainty, below 1,000yrs.

Modern (i.e. post bomb) fresh waters have $^{36}\text{Cl}/\text{Cl}$ in the range 2 to 6 10^{-15} (Phillips, 2000). Surface aquifers (weathered part of the aquifer, 0-30m) have a mean residence time of 15-20yrs and annual renewal rates ranging from 2 to 15% (ref). Thus it might be

expected that these aquifers only contain low amounts of ^{36}Cl . The dotted line in Fig. S1 represents the potential mixing line of modern aquifer waters including various amounts of bomb ^{36}Cl and low Cl content.

Saline samples have ^{36}Cl above those of post-bomb fresh waters and present CFC concentrations below or close to the detection limit. They cannot represent modern aquifer waters including a slight bomb component as they are older than 50yrs and much more saline.

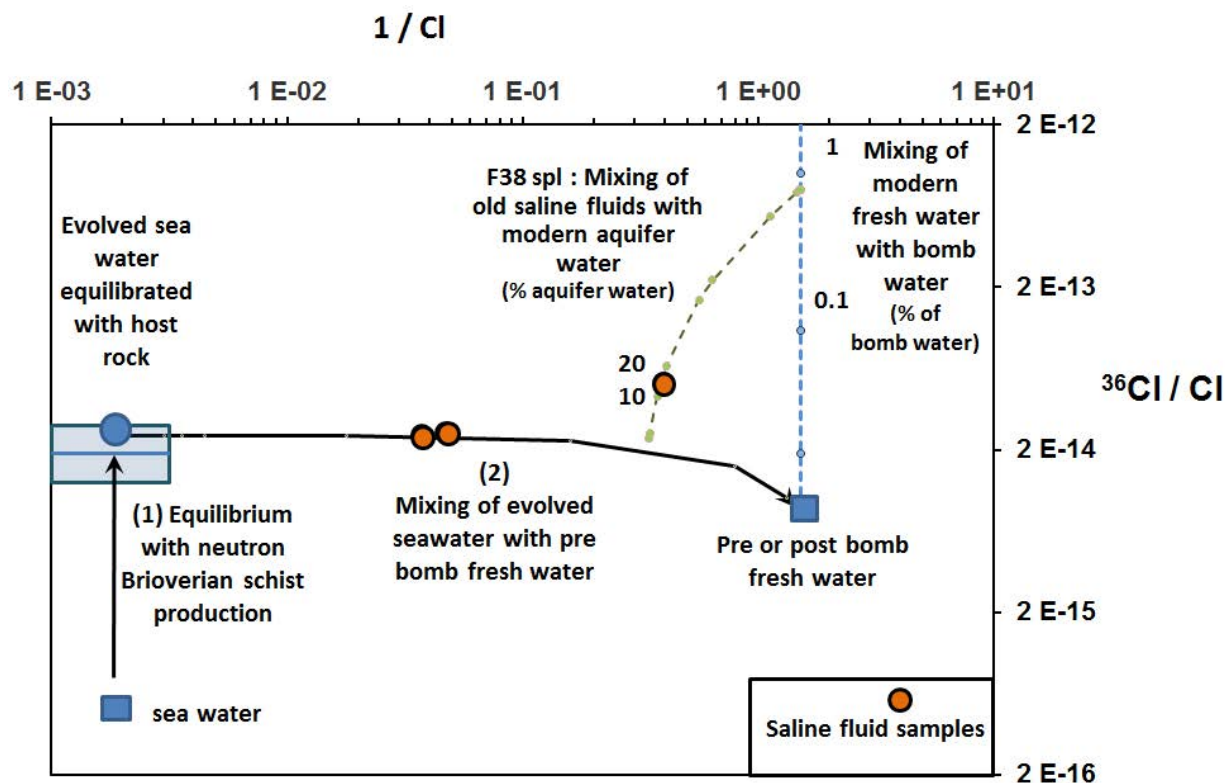


Figure S1. ^{36}Cl -chlorine versus $1/\text{Cl}$

We interpret these samples as resulting from seawater introduction in the basement long-time ago. Seawater has initially a low $^{36}\text{Cl}/\text{Cl}$ (Argento et al., 2010) ratio and then equilibrates with the neutron production flux of the rock (arrow 1 in Figure S1). The secular equilibrium value (Rse) has been defined from the U and Th concentration in the Brioeverian schists of the Armorican basement (Dabard and Peucat, 2001; Dabard, M.-P. and Peucat, 1998; Vignerresse et al., 1989) which contains the saline fluids and represents a large part of the Armorican basement (mean U and Th : 3.4 ± 1.4 and 10.4 ± 2.5 ppm) which provides a Rse range of $1.09 \cdot 10^{-14}$ to $2.16 \cdot 10^{-14}$ and a mean of $1.62 \cdot 10^{-14}$.

The Ploemur schist which is close to a granite has slightly higher U and Th concentrations (9 and 16 ppm) and a Rse of $2.69 \cdot 10^{-14}$. These values are represented in Fig. S1 as the box on the left hand side. The saline sample presents values that are close to the highest range, which suggest that they have reached the secular equilibrium and thus present residence time higher than $1 \cdot 10^6$ yrs.

Then, these fluids are strongly diluted by glacial fluids which are considered as an equivalent of pre-bomb meteoric fresh water. As these fluids only contain very low Cl concentrations, they almost have no effect on the $^{36}\text{Cl}/\text{Cl}$ ratio unless mixing reaches more than 99% of fresh water.

Finally, the higher $^{36}\text{Cl}/\text{Cl}$ ratio of the Ploemur F38 sample might result from the component of modern surface water which is present in this sample. Mixing of evolved paleo-seawater diluted by glacial fluids with modern aquifer groundwater is sketched in Figure S1. $^{36}\text{Cl}/\text{Cl}$ ratio of F38 sample might be explained by introduction of 10 to 20% of modern aquifer groundwater, which agrees with the 15% deduced from CFC and nitrate concentrations (Table S1).

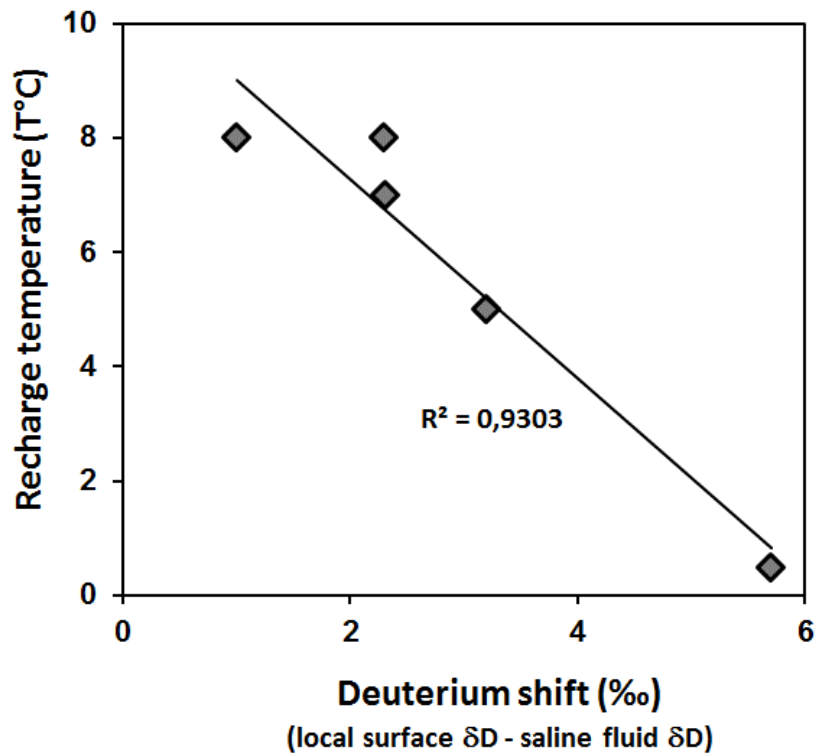


Figure S2. Deuterium shift versus recharge temperature

2.4. Conclusion

Ces travaux mettent en évidence la conservation d'évènements climatiques passés au sein des ressources hydrogéologiques du Massif Armoricaïn. Les signatures de ces évènements climatiques majeurs (transgression marine et période glaciaire) ont été enregistrées par des traceurs environnementaux (Cl⁻, Br⁻, SO₄, gaz nobles, isotopes de l'eau...) et stockées au sein de la ressource en souterraine. La signature marine a principalement été identifiée à partir des concentrations anormalement hautes en chlorure dans les eaux souterraines et est caractéristique d'un climat plus chaud et de l'élévation du niveau de la mer. La seconde phase, d'origine glaciaire est mise en évidence par la signature des gaz nobles et des isotopes de l'eau. Les informations apportées par l'étude de ces divers traceurs environnementaux dans le cas du Massif Armoricaïn sont multiples, puisqu'ils permettent:

- D'identifier les évènements climatiques majeurs qui ont affecté la ressource en eau souterraine (transgression marine et période glaciaire)
- De reconstruire l'histoire hydrogéologique des aquifères de socle du Massif Armoricaïn sur plusieurs millions d'années
- De caractériser le fonctionnement hydrogéologique global durant les différentes phases climatiques et par conséquent de conceptualiser ce fonctionnement
- De contraindre les échelles de temps de circulation des fluides
- D'apporter des informations sur le temps de réponse de ces systèmes à des perturbations majeures
- De montrer la sensibilité des ressources hydrogéologiques aux variations climatiques

Ces signatures climatiques conservées dans la mémoire du système hydrogéologique actuel témoignent donc de l'importance des impacts climatiques sur la ressource en eau souterraine. Au sein du contexte climatique actuel, ce type d'information est primordial puisqu'il souligne la nécessité de s'intéresser à l'impact du changement climatique sur la ressource en eau souterraine. De plus, ces empreintes fournissent de précieuses informations, indispensables à la conceptualisation du fonctionnement hydrodynamique des aquifères du Massif Armoricaïn sur le long terme et ouvrent de nombreuses perspectives de travaux (Chapitre 5).

Chapitre 2. Impacts des variations climatiques passées. Implication dans la compréhension du système hydrogéologique.

Les signaux climatiques répertoriés dans ces deux études sont de très grandes ampleurs, c'est pourquoi ils restent visibles au sein de la ressource en eau souterraine sur de très longues échelles de temps (dizaines de milliers d'années aux millions d'années). Même si dans le cadre du changement climatique les variations climatiques attendues (hausse de la température de 0,3 à +4,8 °C à la fin du siècle et hausse du niveau marin de 26 à 82 cm) sont de moindre ampleur, celles-ci auront un impact sur les ressources hydrogéologiques et se feront ressentir sur des échelles de temps plus restreintes et particulièrement à l'échelle humaine.

Chapitre 2. Impacts des variations climatiques passées. Implication dans la compréhension du système hydrogéologique.

Chapitre 3. Relation eau souterraine – zones humides et variations climatiques

3.1. Introduction

Les compartiments d'eau de surface sont continuellement en interaction avec les eaux souterraines et ceci dans presque tous les types d'environnements. Ces compartiments d'eau de surface sont des ruisseaux, des rivières, des lacs ainsi que par des zones humides font partie intégrante du système hydrogéologique (Winter, 1999). L'intérêt grandissant porté à ces relations depuis plusieurs dizaines d'années s'est concrétisé par la mise en commun des compétences de géologues, d'hydrologues et de biologistes débouchant sur la conceptualisation des interactions entre ces compartiments de surface et l'eau souterraine (Sophocleous, 2002). La conceptualisation des interactions entre l'eau souterraine et chaque compartiment de surface (rivière, lac et zones humides) est brièvement rappelée dans les 3 paragraphes suivants.

3.1.1. Relations eau souterraine et compartiments de surface

Relation eau souterraine – rivière

L'interaction entre l'eau souterraine et la rivière est relative au sens d'écoulement entre ces deux compartiments:

- Les écoulements sont dirigés de l'aquifère vers le réseau hydrographique, ce qui équivaut à une charge hydraulique de l'aquifère supérieure à celle de la rivière. Dans ce cas, le réseau hydrographique est donc alimenté par la nappe sous-jacente (Figure 26, cas A).
- Lorsque le niveau de la nappe est inférieur à celui de la rivière, les écoulements sont orientés dans le sens inverse (du réseau hydrographique vers l'aquifère).

Chapitre 3. Relation eau souterraine – zones humides et variations climatiques

Dans ce cas, le réseau hydrographique alimente la nappe sous-jacente, cependant ces écoulements sont dépendants de nombreux paramètres (profondeur de la nappe, conductivité hydraulique du milieu...). Dans cette configuration, la rivière et la nappe peuvent être connectées (Figure 26, cas B) ou bien déconnectées et séparées par une zone insaturée (Figure 26, cas C). Les entre les cours d'eau et les aquifères sont diverses et varient dans le temps et dans l'espace. Ces échanges peuvent aussi être en fonction des variations saisonnières. En effet, en période d'étiage, le débit des rivières n'est plus alimenté par le ruissellement, l'alimentation ne se fait donc que par les eaux souterraines. Il est ainsi possible d'observer l'inversion des échanges selon la saison.

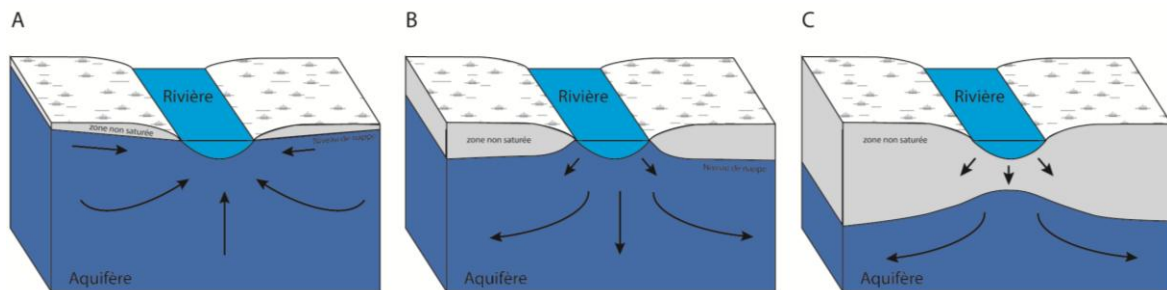


Figure 26. Relation eau souterraine – rivière. Cas A : Écoulements ascendants (aquifères vers rivière). Cas B : Écoulements descendants (Rivière vers aquifère) avec connexion. Cas C : Écoulements descendants (Rivière vers aquifère) sans connexion.

Relation eau souterraine – lac

Le régime hydrologique d'un lac est fortement dépendant du système d'écoulement régional auquel il appartient. L'alimentation d'un lac par les eaux souterraines est contrôlée par les caractéristiques du bassin versant (topographie) ainsi que par l'environnement hydrogéologique (Gedeon, 1999). Les interactions entre l'eau souterraine et les lacs peuvent se résumer en 3 grands cas (Figure 27) :

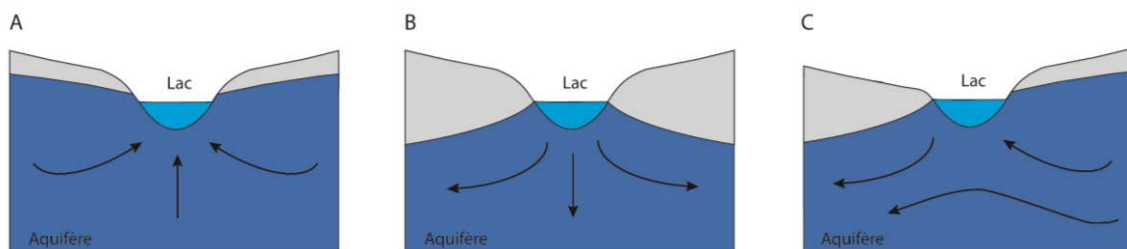


Figure 27. Relation eau souterraine – lac. Cas A : Écoulements ascendants (aquifères vers lac). Cas B : Écoulements descendants (lac vers aquifère). Cas C : Cas mixte.

Relation eau souterraine – zones humides

Les interactions existantes entre les eaux souterraines et les zones humides sont dépendantes des conditions géologiques locales. En effet, l'interaction entre la zone humide et l'aquifère sous-jacent dépend de l'existence ou non d'une formation les séparant, mais aussi de la perméabilité de cette formation (Figure 28):

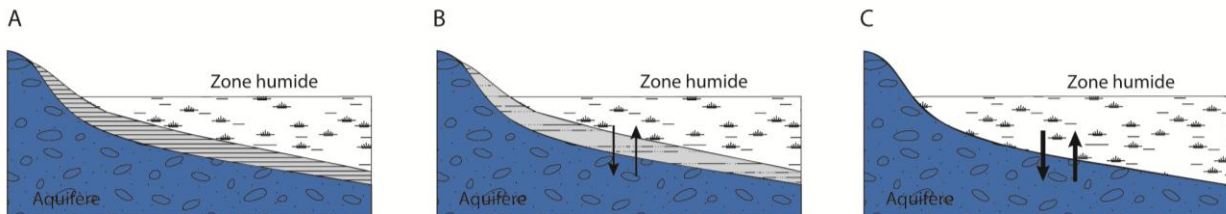


Figure 28. Relation eau souterraine – zone humide. Cas A : Pas d'échanges, l'aquifère et la zone humide sont déconnectés. Cas B : Échanges possibles, mais dépendants de la perméabilité de la formation les séparant. Cas C : Échanges actifs.

Dans le 1^{er} cas (Figure 28, Cas A), la zone humide est séparée de l'aquifère par une zone imperméable (comme des argiles), empêchant les échanges. Dans le 2^{ème} cas (Figure 28, Cas B), les échanges entre les deux compartiments sont limités par une formation peu perméable. Enfin dans le 3^{ème} cas (Figure 28, Cas C) l'interaction est forte du fait de l'absence de formation entre la zone humide et l'aquifère. Enfin, ces interactions sont aussi tributaires du niveau relatif de la nappe et de la zone humide. L'écoulement étant ascendant dans le cas où le niveau de la nappe est supérieur à celui de la zone humide (zone humide alimentée par la nappe) et inversement. Les interactions entre les zones humides et les diverses unités hydrologiques ont été le sujet de nombreuses études, soulignant notamment la recharge en eau souterraine de ces environnements (Bendjoudi et al., 2002; Cey et al., 1998; Hunt et al., 1999, 1996; Weng et al., 2003; Winter, 1999).

Les interactions entre les eaux souterraines et les différents compartiments d'eau de surface sont complexes. Les interactions entre les lacs, les rivières et les zones humides avec les eaux souterraines déterminent la répartition des écoulements au sein du système souterrain entre les systèmes de différents ordres (régionaux et locaux (Toth, 1962)), et dépendent des propriétés géologiques et des conditions climatiques (Sophocleous, 2002; Winter, 1999, 1995). Ces échanges permanents entraînent que l'impact d'une perturbation qu'elle soit climatique ou anthropique sur l'un des

composants du système hydrogéologique (rivières, lacs, zones humides, aquifère...) se répercutera sur le fonctionnement de l'ensemble du système.

3.1.2. Activités anthropiques

Or, pour satisfaire ses besoins, l'homme puise de grandes quantités d'eau à partir des compartiments de surface (rivières, lacs...) ainsi qu'à partir des aquifères. Dans certaines régions, pour compenser l'insuffisance des eaux de surface l'utilisation de la ressource en eau souterraine est requise. Or, l'exploitation de l'eau souterraine peut dans certains cas se caractériser par une surexploitation de la ressource (Gleeson et al., 2012; Scanlon et al., 2012a, 2012b; Taylor et al., 2013; Voss et al., 2013) et avoir des effets dévastateurs sur le débit des rivières, l'alimentation en eau des zones humides et les écosystèmes associés (Aeschbach-Hertig and Gleeson, 2012; Foster and Chilton, 2003). Durant les dernières décennies, les flux d'eau ont été impactés par l'homme avec une ampleur sans précédent. Ces empreintes sur la ressource en eau caractérisées par des modifications de la connectivité et/ou de l'altération des processus d'échanges entre les compartiments (Sophocleous, 2002) se reflètent sur les réservoirs d'eau de surface (rivières, lacs, zones humides...) et souterrains.

3.1.3. Changement climatique

En plus de la demande croissante en eau, le changement climatique et sa variabilité ont un effet sur le cycle de l'eau et par conséquent se répercutent sur sa disponibilité (Arnell, 1999, 2004; Dai, 2012; Haddeland et al., 2011; Sheffield et al., 2012). Un intérêt grandissant s'est porté sur le fait que le comportement « naturel » du climat subit des perturbations. Le réchauffement global, observé particulièrement sur les deux dernières décennies est attribué en grande partie aux activités humaines et à l'émission de gaz à effet de serre dans l'atmosphère (Cubash et al., 2013; IPCC, 2007).

Dans cette optique, plusieurs modèles de circulation à l'échelle globale GCMs (« General Circulation Models ») ont vu le jour pour simuler le climat en fonction de l'évolution des concentrations en gaz. Les GCMs, outils indispensables pour la simulation des variables climatiques, se définissent comme des modèles numériques qui s'efforcent de simuler le climat global en calculant l'évolution de l'atmosphère en trois dimensions basée sur les lois de conservation de masse, de dynamique, d'énergie et de vapeur d'eau

pour l'atmosphère (Grotch and MacCracken, 1991). Un GCM est généralement le résultat du couplage de plusieurs modèles : circulation atmosphérique globale, circulation océanique, glace-mer, processus de surface, processus de la biosphère, cycle du carbone, etc. (IPCC, 2007). Ces GCMs fournissent donc des variables climatiques à très grande échelle, typiquement de 2° de latitude à 5° de longitude. Néanmoins, les études hydrologiques se font à des échelles plus restreintes (quelques km² à quelques centaines de km²), les sorties des GCMs ne sont donc pas directement adaptées dans le cadre de ces études, du fait de la non-prise en compte des effets locaux comme l'utilisation des sols, l'hydrologie, la topographie, etc..

C'est la raison pour laquelle de nombreuses techniques de « downscaling » ont été développées dans le but de rendre utilisable les variables climatiques à une échelle plus locale (Fowler et al., 2007). Les RCMs (« Regional Climate Models ») issus du « downscaling » possèdent une résolution plus fine (de l'ordre de 0.5° de latitude et de longitude) et sont plus performants dans la reproduction du climat régional (et des événements extrêmes) du fait de la prise en compte de caractéristiques régionales supplémentaires, comme la végétation, les compartiments d'eau de surface, la topographie, etc. Les RCMs sont donc utilisés pour la simulation des variables climatiques nécessaires aux études d'impact du changement climatique sur les ressources en eau en fonction de scénarios d'émissions de gaz à effet de serre.

L'IPCC, Intergovernmental Panel on Climate Change a développé une gamme de scénarios d'émissions (Rapport Spécial sur les Scénarios d'Emissions, SRES scenarios), regroupés en 4 grandes familles, qui envisagent des scénarios d'émissions en fonction d'un large panel de facteurs économiques, démographiques, technologiques, etc. (Nakicenovic et al., 2000). Les principales caractéristiques sont décrites dans le rapport spécial, la Figure 29 présente les projections d'émissions mondiales de gaz à effet de serre (CO₂, CH₄, N₂O et gaz fluorés) pour les 4 grandes familles et 6 groupes de scénarios pour la période de 2000 à 2100. Ces scénarios d'émissions de gaz à effet de serre peuvent être utilisés dans les GCMs pour produire des scénarios de changement climatique.

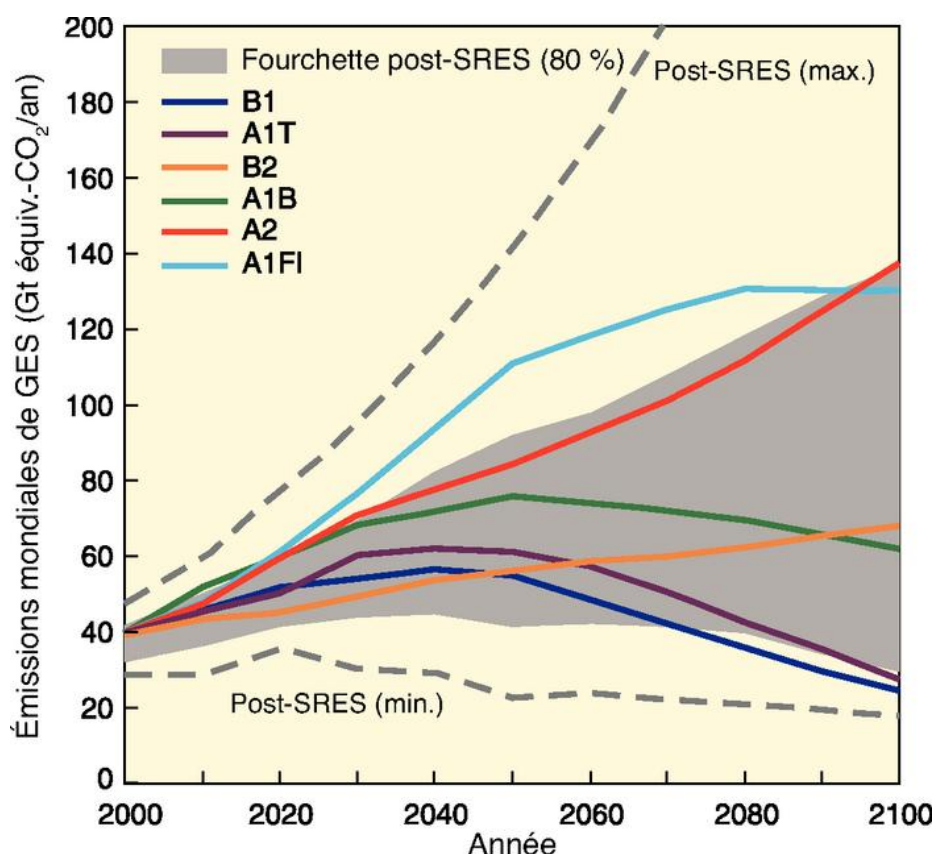


Figure 29. Émission mondiale de gaz à effet de serre pour toutes les sources (énergie, industrielle...) sur une période de 2000 à 2100 en l'absence de politiques climatiques additionnelles (exprimée en Gigatonnes équivalent de CO₂/an). Les lignes colorées correspondent aux 6 scénarios illustratifs de référence.

Les études sur l'impact du changement climatique sur la ressource en eau sont souvent restreintes aux eaux de surface et/ou négligent les eaux souterraines (Singh and Kumar, 2010), bien que celles-ci fassent partie intégrante du système hydrologique. La compréhension de ces interactions est un élément fondamental pour la gestion des ressources en eau qui y sont associées, d'où depuis quelques années une émergence de modélisation numérique de ces systèmes par couplage des scénarios de changement climatique avec des modèles hydrologiques (Bradley, 2002; Frei et al., 2010; Grapes et al., 2006; Lischeid et al., 2010; Reeve et al., 2001, 2000; van Roosmalen et al., 2009; Wilsnack et al., 2001).

Dans le but de pérenniser la ressource, l'étude et la gestion des ressources en eau associées à ces environnements, nécessitent de prendre en compte les diverses activités anthropiques pouvant perturber le système ainsi que les variations climatiques pouvant survenir au cours du temps.

L'étude suivante menée durant cette thèse s'inscrit dans cette thématique visant à évaluer et à comparer l'impact des variations climatiques et des activités anthropiques sur l'étendue des zones humides et le stock d'eau souterraine à partir d'un exemple dans le Parc Naturel Régional des Marais du Cotentin. Les zones humides au sein de ce parc couvrent environ 30000 ha dont les caractéristiques écologiques ont été reconnues aux titres de directives européennes et de la convention de Ramsar. À ce riche écosystème s'ajoute la présence d'une ressource en eau souterraine potable d'intérêt régional. Dans l'optique de pérenniser les atouts de ce territoire ainsi que toutes les activités y prenant place, plusieurs enjeux ont été identifiés : (1) préserver et conforter la biodiversité (2) assurer une gestion équilibrée de la ressource en eau (3) encourager un écodéveloppement (4) valoriser les différents patrimoines comme facteur d'attractivité et (5) cultiver la démocratie participative pour mieux mettre en œuvre le projet de territoire. Or, ces zones humides ont déjà été soumises à de nombreuses pressions (drainage pour l'agriculture, pompages, exploitation de tourbe) auxquelles viennent s'ajouter aujourd'hui les problématiques associées au changement climatique. C'est dans ce contexte qu'a été initiée l'étude qui suit pour répondre à une demande du Parc Naturel Régional et plus particulièrement à certaines questions :

- Comprendre le fonctionnement hydrologique de l'ensemble du système, notamment la mise en évidence des interactions entre les zones humides et la nappe souterraine sous-jacente.
- Appréhender l'impact des pompages AEP à l'heure actuelle ainsi qu'à court terme, sur les zones humides. Et dans un contexte plus global, mesurer l'impact des activités anthropiques sur celles-ci.
- Envisager et prédire l'impact du changement climatique sur les zones humides.

Les interactions existantes entre l'aquifère et les compartiments de surface ont été mises en évidence tout en prenant compte les paramètres déterminants tels que la topographie, les propriétés géologiques du milieu et l'environnement climatique. Les résultats obtenus à partir du modèle numérique permettent d'établir un schéma du bilan hydrologique et de comprendre les interactions entre les différents compartiments (Figure 30). Les différents échanges ont été calculés à partir du volume d'eau (en m³) passant d'un compartiment à un autre sur une année et représentés sous forme de pourcentage par rapport à la recharge totale (~ 51 Mm³). Les résultats montrent

clairement que l'aquifère alimente les zones humides avec des échanges 5 fois plus important de l'aquifère vers la zone humide. D'autre part, ces résultats mettent aussi en avant l'importance des pompages au sein de la tourbière, pompages qui semblent constituer un des exutoires les plus importants de ce bassin (environ 1/5 de la recharge totale et deux fois plus que les pompages).

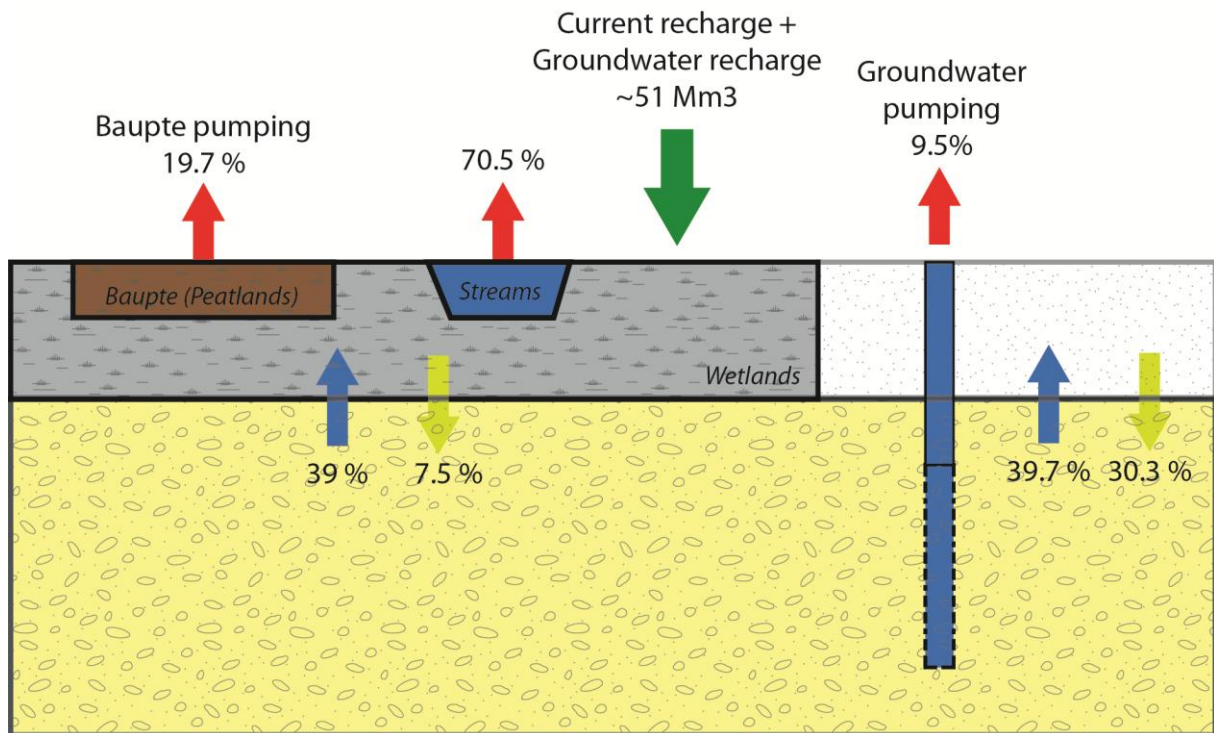


Figure 30. Bilan hydrologique

Le modèle numérique édifié permet d'investiguer l'impact des variations climatiques et des activités anthropiques sur l'étendue des zones humides et de proposer des solutions de gestion dans le but de pérenniser le bon fonctionnement du système hydrogéologique.

L'article s'organise comme suit : après une introduction (section 1), le site d'étude et ses caractéristiques sont présentés (section 2). La section 3 présente le modèle numérique et les différents scénarios testés. Les résultats de la sensibilité des zones humides aux différentes perturbations (climatiques, activités anthropiques) sont présentés en section 4. La section 5, quant à elle propose une discussion sur les limites et les incertitudes du modèle, l'impact des perturbations tout en proposant des solutions de gestion et se termine par une mise en évidence des impacts passés.

3.2. *Investigating the respective impacts of groundwater exploitation and climate change on wetland extension over 150 years*

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Investigating the respective impacts of groundwater exploitation and climate change on wetland extension over 150 years



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SUMMARY

Peatlands are complex ecosystems driven by many physical, chemical, and biological processes. Peat soils have a significant impact on water quality, ecosystem productivity and greenhouse gas emissions. However, the extent of peatlands is decreasing across the world, mainly because of anthropogenic activities such as drainage for agriculture or groundwater abstractions in underlying aquifers. Potential changes in precipitation and temperature in the future are likely to apply additional pressure to wetland. In this context, a methodology for assessing and comparing the respective impacts of groundwater abstraction and climate change on a groundwater-fed wetland (135 km²) located in Northwest France, is presented. A groundwater model was developed, using flexible boundary conditions to represent surface–subsurface interactions which allowed examination of the extent of the wetland areas. This variable parameter is highly important for land management and is usually not considered in impact studies. The model was coupled with recharge estimation, groundwater abstraction scenarios, and climate change scenarios downscaled from 14 GCMs corresponding to the A1B greenhouse gas (GHG) scenario over the periods 1961–2000 and 2081–2100. Results show that climate change is expected to have an important impact and reduce the surface of wetlands by 5.3–13.6%. In comparison, the impact of groundwater abstraction (100% increase in the expected scenarios) would lead to a maximum decrease of 3.7%. Results also show that the impacts of climate change and groundwater abstraction could be partially mitigated by decreasing or stopping land drainage in specific parts of the area. Water management will require an appropriate compromise which encompasses ecosystem preservation, economic and public domain activities.

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1. Introduction

Peatlands are complex and fragile ecosystems driven by many physical, chemical, and biological processes. Numerous studies have provided a comprehensive understanding of wetland hydrology, especially regarding the interactions between surrounding aquifers and surface water networks (Bradley, 2002; Frei et al., 2010; Grapes et al., 2006; Lischeid et al., 2010; Reeve et al., 2000; van Roosmalen et al., 2009; Wilsnack et al., 2001; Winter, 1999). Because peat soils can serve as sinks, sources, and transformers of nutrients and other chemical contaminants, they have a significant impact on water quality, ecosystem productivity and greenhouse gas emissions (Hemond and Benoit, 1988; Johnston,

1991; Kasimir-Klemedtsson et al., 1997; Roulet, 2000). The extent of peatlands is tending to decrease worldwide, (Estimated to 6% over the period 1993–2007 – Prigent et al. (2012)). However, peatlands are considered as important carbon reserves (15–30% according to Botch et al. (1995), Turunen et al. (2002)), and important potential sources of CO₂ even though they cover only 3–4% of emerged areas on the earth. As the oxygen concentration in peat increases due to water drawdown, surface decomposition is enhanced by bacterial aerobic processes (Holden et al., 2004). Oxygen enhances organic matter mineralization, leading to CO₂ release to the atmosphere and nutrients production, particularly carbon-bound nitrogen and sulfur. Decreasing groundwater levels can also cause land subsidence, due to the reorganization of the peat structure (Silins and Rothwell, 1998).

The hydrology of the peat layer and extent of this peat area are impacted by drainage for agriculture, groundwater abstractions in underlying aquifers and climate change. The general impact of climate change on hydrological systems has been studied, focusing on surface water (Christensen et al., 2004; Fowler et al., 2007a),

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and more recently on groundwater reserves (e.g. Goderniaux et al., 2009, 2011; Green et al., 2011; Herrera-Pantoja and Hiscock, 2008; Holman et al., 2011; Scibek et al., 2007; Woldeamlak et al., 2007). However, few studies have addressed the impact of climate on the evolution of peatlands, which are specific ecosystems located at the interface between surface water and groundwater. Moreover, the respective impacts of climate change and anthropic water abstraction on wetlands have not been investigated and compared.

Peatlands are commonly observed in lowland areas where shallow gradients, impermeable substrates or topographic convergence maintain saturation. Peatland classification is generally related to two fundamental factors: source of nutrients and source of water. Bogs or ombrotrophic peatlands are dependent on precipitation for water and nutrient supply, whereas minerotrophic peatlands or fens rely on groundwater (Johnson and Dunham, 1963). As a consequence, two different points of view have generally been adopted in studies of the impact of climate change. Thompson et al. (2009) performed an impact study on the Elmley marshes (8.7 km²) in England using a coupled surface–subsurface model, where subsurface is represented by a single uniform layer. In their study, precipitation and evapotranspiration were the main hydrological processes, due to the impoundment of the marshes within embankments and their low hydraulic conductivity. Conversely, other studies emphasized the importance of the interactions with groundwater. Candela et al. (2009) developed a groundwater model (415 km²) for a basin in the Island of Marjorca (Spain), to assess the impact of climate change on groundwater resources and on springs discharging into a smaller wetland area. Herrera-Pantoja et al. (2012) used a generalized groundwater model of eastern England wetlands to assess climate change impacts on water levels and their consequences on typical plant species. Barron et al. (2012) assessed the risks for wetlands and groundwater-dependent vegetation in the southern half of the Perth Basin (~20,000 km², Australia) under future climate change scenarios. Their study is based on a global approach using coefficients of groundwater sensitivity to climate change, and a regional-scale groundwater model.

In this study, we considered peatlands as components of a complex system where the different surface and subsurface compartments interact. Our general objective was to evaluate and compare the competing impacts of climate change and water abstraction activities on groundwater storage and the extents of wetland areas. We focused on a 135 km² peatland area in the Cotentin marshes (Northwest France). Our three main objectives were: (i) to understand surface–subsurface connectivity and associated wetland hydrological sensitivity, (ii) to quantify the impact of projected increases in groundwater abstraction, and (iii) to estimate the impact of climate change at the end of this century. These objectives have been attained by using a 3D groundwater model for the Cotentin wetland area.

2. Study area

2.1. The Cotentin marshes

The Cotentin marshes are located within a large watershed in Normandy (Northwest France, see Fig. 1). The study area is situated within a natural reserve, and extends over approximately 135 km². Topography ranges from 0 to 30 m above sea level. Mean annual precipitation and potential evapotranspiration for the period 1946–2010 (from two climatic stations, Fig. 1) were 910 mm/yr and 630 mm/yr, respectively. In the lowland areas, the vast wetlands and peatlands partly consist of peat soils and are located along 3 main rivers: the 'Sèves' in the North, the 'Holerotte' in the West, and the 'Taute' in the South (Fig. 1). As suggested by hydrologic fluxes and chemical features (Auterives et al., 2011), this wetland area is closely related to groundwater. It is connected with an underlying highly transmissive aquifer and surface-water bodies are integral parts of the groundwater flow systems. For several centuries, this large wetland has undergone numerous disturbances. In the 18th century the wetland was flooded 9 months per year (Bouillon-Launay, 1992). Since 1712, a human-controlled drainage system has gradually been set up. From 1950 until now, the flooding season has been reduced to only 3 months on average

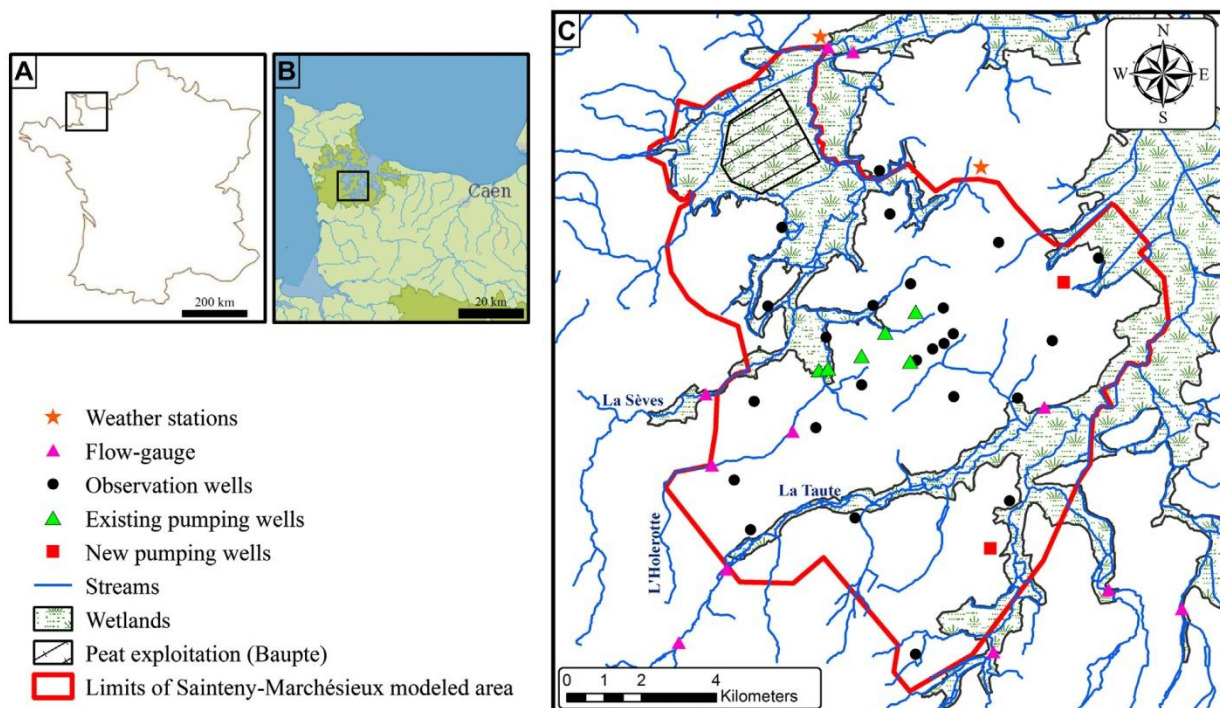


Fig. 1. Location of the Saintenty-Marchésieux basin. (A) Map of France. (B) Map of the Cotentin region. (C) View of the modeled area.

due to agricultural constraints. The top peat profile is thus subjected to longer periods of desiccation. Beside agricultural pressure, the underlying aquifer is also used as a drinking water supply, since 1992. Due to an increasing demand for high-quality water, the authorities plan to increase groundwater abstraction in the near future. The Cotentin peatlands are nevertheless also classified as a natural reserve for specific wildlife and plant species. Additionally, geotechnical perturbations such as the collapse of parts of houses or fissures in constructed walls have also been reported along the border of the peatland. These perturbations have generated public manifestations, and the filing of legal claims in early 2012. It was often claimed during these manifestations that groundwater extraction was responsible for the observed damage. As a consequence, the Cotentin marshes are at the center of different interests, which must be integrated by stakeholders into their management plans: ecological activities through the preservation of wetlands, economic activities through the preservation of farmland, and public domain activities through the distribution of drinking water.

2.2. The Sainteny-Marchésieux aquifer

The geology of the Sainteny-Marchésieux study area, located in the Cotentin marshes, corresponds to a graben structure (Baize, 1998), bounded by NE-SW and NNW-SSE faults (Fig. 2), with a depth of 150 m. The substratum is considered as impermeable and corresponds to Precambrian geological formations to the south and west and Permo-Trias to the east and the north (Fig. 2). Within the graben structure, two different aquifer areas can be distinguished (Fig. 2). (1) The Sainteny aquifer in the northwest extends over approximately 35 km². It consists of shelly sands up to 100 m thick, characterized by relatively high hydraulic conductivity. (2) The Marchésieux aquifer, in the south, extends over approximately 100 km². This area is characterized by different lithologies, including sandstones, shales and sandy loams. These formations have a

maximum total thickness of 150 m and are considered less permeable than the shelly sands of the Sainteny aquifer. These thick formations are overlain by (1) Holocene peats, ranging in thickness from 1 to 10 m in the wetland area (Fig. 1) and (2) by sands (up to 10 m) elsewhere. According to the observed groundwater heads and the hydraulic conductivity of these lithologies, the aquifer is considered as confined below the peatlands and unconfined below the sands.

Generally, groundwater flows from southwest to northeast and the aquifer is drained by a dense hydrographic network. High and low areas act as recharge and discharge zones, respectively, (as conceptually shown in Fig. 3). Currently, groundwater is predominantly abstracted in the Sainteny aquifer, with about 5 million m³ pumped each year in 5 different existing wells (Fig. 1). The Marchésieux aquifer exploitation is limited to a single existing well, with a pumping rate of about 0.14 million m³ per year. Groundwater abstraction represents approximately 9% of the total recharge rate.

In the north of the catchment, the peatlands in the 'Baupte' area (Fig. 1) were exploited from the 1950s to 2006. Peat extraction implied a considerable lowering of the water table. Currently, a large pit of about 0.4 km² remains and the average water level in this zone is artificially maintained about 4.9 m below the ground surface to avoid flooding of certain areas of farmland.

3. Modeling

3.1. Model implementation

The 3D groundwater model has been developed with the Modflow 2005 finite difference code (Harbaugh, 2005). The modeled area corresponds to the Sainteny-Marchésieux hydrogeological catchment which is globally defined by the limits of the graben structure. Boundary conditions along the model limits have been implemented as follows (Fig. 2):

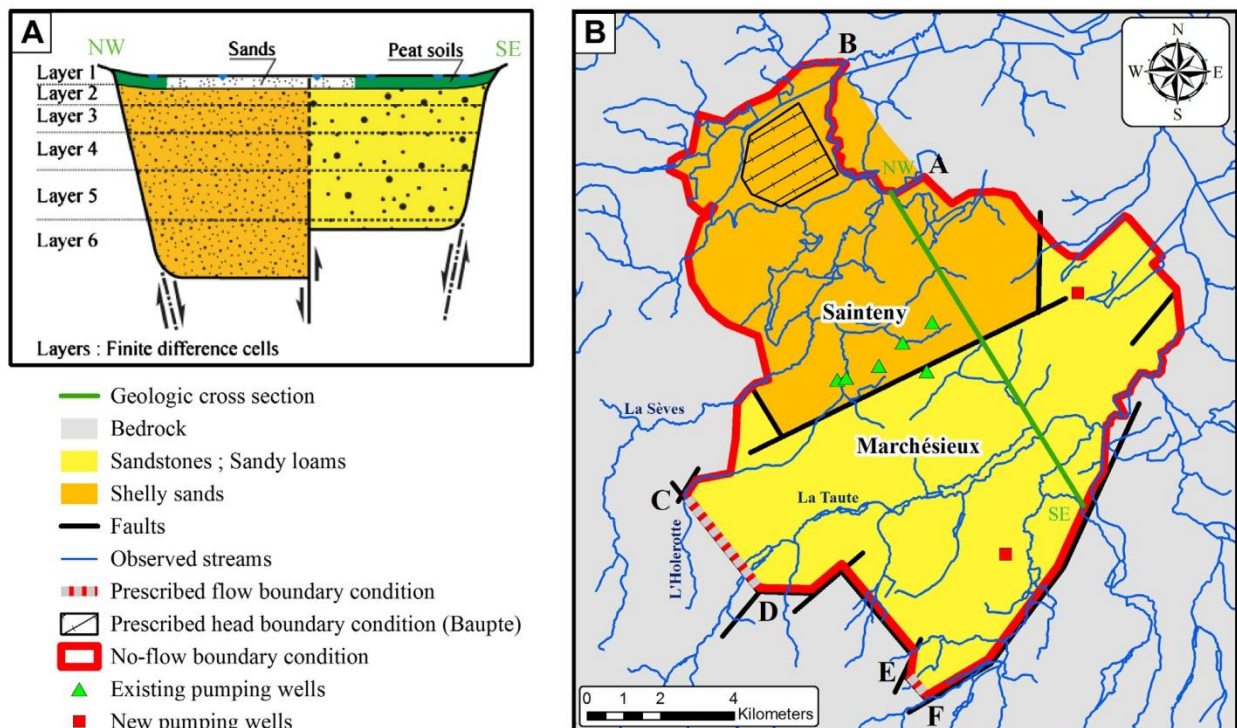


Fig. 2. Geology of the Sainteny-Marchésieux basin and boundary conditions of the model. (A) Geologic cross section. (B) Map of boundary conditions of the model.

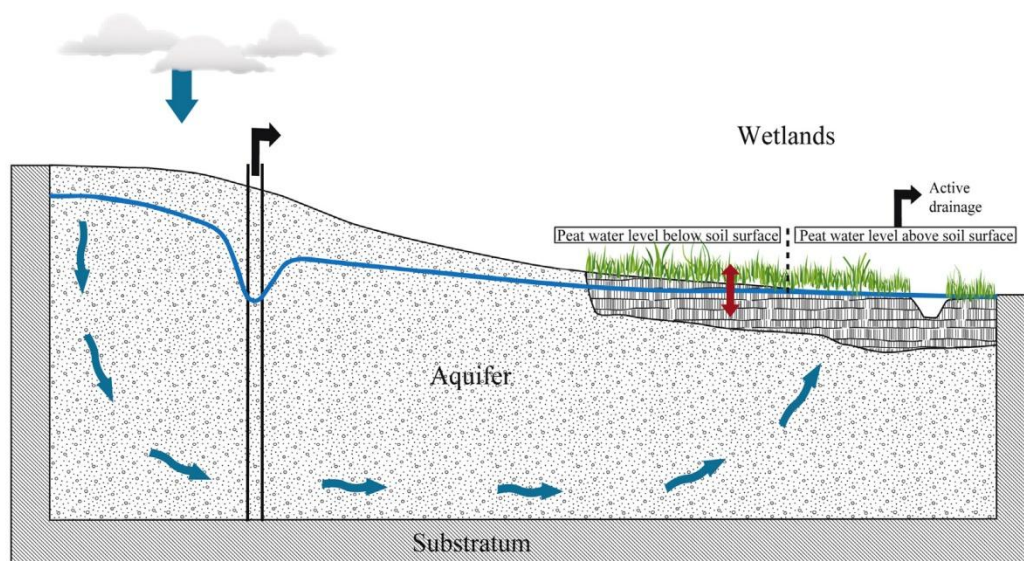


Fig. 3. Conceptual model.

- Sections B–C, D–E and F–A correspond to the interface between bedrock and sediments inside the graben. According to the geology and measured groundwater levels, this interface is considered as impermeable. A no-flow boundary condition is prescribed along these sections.
- Section A–B corresponds to a stream section, which is considered as a main drainage divide. A no-flow boundary condition is implemented along this stream section.
- Sections C–D and E–F also correspond to the graben limits but measured groundwater levels show that groundwater fluxes, from the adjacent geological formation, feed the aquifer. Along these sections, a groundwater flux, equal to the recharge rates times the upstream areas, is prescribed.

Inside the modeled area, a seepage boundary condition (head dependent flux – ‘Drain package’) is applied at the ground surface. This boundary condition enables groundwater to leave the system only when the simulated hydraulic head is above the topographic surface, according to a conductance coefficient. This type of boundary condition is particularly useful in this wetlands context, where the extent of the discharge areas is dependent on recharge rates. Fluxes abstracted for drinking water distribution are applied to the nodes corresponding to the pumping wells (see Section 3.5.1). Finally, a prescribed head boundary condition is applied to the Baupte peatland extraction area (0.4 km²) where the water level is artificially maintained at 4.9 m below the ground surface. This boundary condition can be used in this circumstance because the calculated heads are never lower than the bottom of the peat exploitation. The bottom of the model is considered as impermeable, and implemented with a no-flow boundary condition.

3.2. Model discretization

The study area was discretized using 90 by 90 m cells and 6 layers, with a total number of approximately 100,000 cells. The top of the first layer corresponds to the topographic surface, extracted from a digital elevation model of the region. This first layer is 10 m thick and corresponds to the quaternary peats and upper sands. The interface between the first and second layer corresponds to the top of the aquifer which is composed of shelly sands, sandstones and sandy loams (see Section 2). The depth of the aquifer base has been defined from borehole data and ranges from 70 m

to 150 m below sea level. Five horizontal finite difference layers are used to represent this aquifer.

3.3. System stresses

Recharge and wells pumping rates are applied as input to the model. Pumping rates are calculated from the abstracted groundwater volumes, which have been collected for years by the regional water agencies. Recharge values are applied on the whole modeled area. They are computed externally using a water balance method, based on a modified version of the parsimonious monthly lumped model GR2M (Mouelhi et al., 2006). The GR2M model obtained one of the best performances in a benchmark test of 410 basins throughout the world in different climatic contexts, as compared to 9 other models with generally more parameters (Mouelhi et al., 2006). The GR2M model has been designed to separate rainfall into actual evapotranspiration, surface runoff and transfer to the routing store, which is interpreted here as aquifer recharge (see model description at <http://www.cemagref.fr/webgr/Index-GB.htm>). Observed rainfall and potential evapotranspiration, provided by ‘Meteo France’, are used as GR2M inputs. The GR2M model is calibrated to monthly surface runoff data, which are calculated by baseflow separation from measured river flow rate time series. Data are available over a time frame ranging from January 1999 to December 2000 and January 2003 to December 2007 and includes both wet (1999–2000) and very dry years (2003–2004), which maximizes the descriptive ability of the model over a large interval of climatic fluctuations. This is particularly important in the context of future climate change where applied stresses typically go beyond the calibration interval. The calibration is limited to a 1-parameter calibration process, which is here the soil storage capacity, and carried out on the square root of surface runoff to allow equal weight to high and low flow situations (Oudin et al., 2006). In the optimization process, the Nash–Sutcliffe criterion (Nash and Sutcliffe, 1970) was used as the objective function, and supplemented with the constraint to conserve the total amount of surface runoff ($\sum Q_{obs} / \sum Q_{sim} = 1$), where Q_{sim} and Q_{obs} are simulated and observed surface runoff (Fig. 4). The Nash–Sutcliffe criterion is 0.70 and the calculated annual recharge ranges from 164 mm/yr to 338 mm/yr. On an annual basis, the total amount of water in rivers is also preserved. The sum of simulated surface runoff and recharge is very close to the observed flow rate

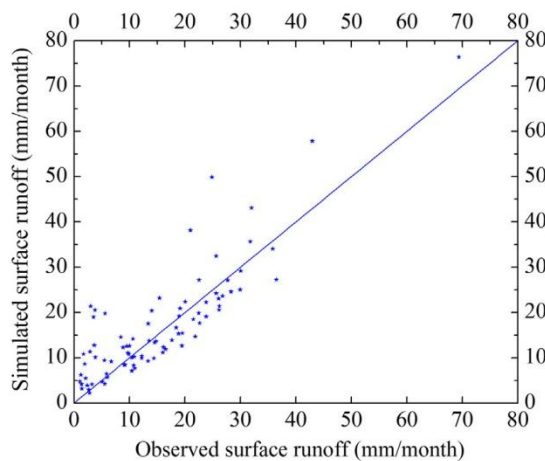


Fig. 4. Monthly surface runoff observed and simulated by the modified version of GR2M model.

in rivers. For the wet (1999–2000) and dry (2003–2004) years, the error is equal to 2% and 4%, respectively. Checking this relation prevents under or overestimation of calculated annual recharge due to errors on the actual evapotranspiration term. This calibrated ‘GR2M’ mass-balance model is subsequently used to externally calculate the recharge to be applied as input to the hydrological Modflow model, for historic and future climatic scenarios (see Section 3.5).

3.4. Calibration and validation of the hydrological model

The hydrological Modflow model was calibrated to the observed (i) aquifer hydraulic heads, (ii) spatial distribution of wetland area and (iii) stream base-flow. The calibration was performed in steady state conditions for two humid and dry contrasted years, 1999–2000 ($R = 338$ mm/yr) and 2003–2004 ($R = 164$ mm/yr), respectively, for which daily climatic data were available. The calibration was performed automatically using the PEST module coupled with Modflow, and by adjusting the hydraulic conductivities of the different geological formations within specific ranges provided by field tests (Auterives, 2007; Auterives et al., 2011). This calibration was validated using data from the hydrologic year 2006–2007 ($R = 263$ mm/yr), which is close to the 1961–2000 average precipitation and temperature statistics (where $R = 250$ mm/yr). Results of the calibration are shown in Table 1, Figs. 5 and 6, for hydraulic conductivities, groundwater levels and wetland surface, respectively. Table 1 shows the calibrated hydraulic conductivities of the geological formations represented in the model. The seepage conductance is set to a high value, calculated from a hydraulic conductivity which is significantly higher than the hydraulic conductivity of the geological layers. Fig. 5 presents residuals for the groundwater levels, calculated as the difference between the observed and simulated values. Fig. 6 shows the observed and simulated wetland areas. The “observed

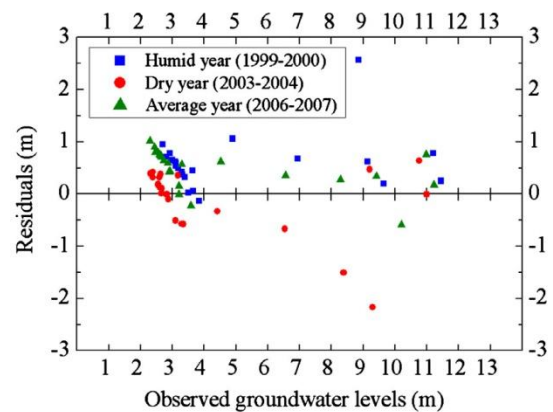


Fig. 5. Residuals for groundwater levels.

wetland areas” are given by cartographic data (data base from the local conservatory park: “Parc Naturel des marais du Cotentin et du Bessin”) which are representative of the mean climatic conditions of the time period 1961–2000. These wetlands are defined as zones where a groundwater level close to the soil surface is maintained during a large part of the year. The “simulated wetland areas” are calculated from the model outputs. A finite difference cell is considered as part of the wetland area when the simulated groundwater level is less than 0.5 m below the ground surface. Observed groundwater discharge volumes are calculated as the difference between the main stream inlets (Sèves, Holerotte and Taute) and outlets (Sèves and Taute) volumes, where gauging stations are located (Fig. 1). The errors between observed and simulated volumes of groundwater drained from the aquifer to the surface domain (through the seepage boundary condition) are below 5%. The volume of water extracted by the prescribed head boundary condition is equivalent to the quantity of water pumped by the Baupré peatland manager: approximately 10 million m^3 each year.

3.5. Future scenarios

Future scenarios for groundwater abstraction and climate change were applied as input to the calibrated model, to assess their respective impacts on the catchment, with particular focus on wetland extension. These scenarios are compared with a reference simulation corresponding to the average recharge and groundwater abstraction for the period 1961–2000. The reference recharge is 250 mm/yr and the abstracted groundwater volumes are 5 million m^3 /yr. The climate change, groundwater abstraction and management scenarios considered in this study are summarized in Table 2.

3.6. Future groundwater abstraction

Groundwater abstraction volumes that will be required in the future are defined from projections made by the local water agency. Four different scenarios are tested, considering an increase in groundwater demand of 10%, 20%, 50% and 100%, relative to the current volumes (5 million m^3 /yr) (Table 2). Two different management plans are considered and tested with the model. The first plan consists in applying the increase in groundwater demand to the pumping rates of the 6 existing wells currently used (Fig. 1) (Scenarios 1–4 in Table 2). The second plan consists in using two new wells located in the Marchésieux aquifer (Fig. 1) to support the increase in groundwater demand (Scenarios 5–8). In this second plan, the abstraction flow rates in the existing wells are kept constant.

Table 1
Calibrated horizontal and vertical hydraulic conductivities.

Layer	Lithologie	K_{xy} (m/s)	K_z (m/s)
Layer 1	Sands	1×10^{-6}	1×10^{-6}
	Peats	8×10^{-7}	7×10^{-8}
Layer 2, 3, 4, 5 and 6	Shelly sands	8×10^{-3}	8×10^{-3}
	Sandstones	8×10^{-4}	8×10^{-4}
	Sandy loams	5.5×10^{-6}	5.5×10^{-6}
	Sandy loams	2×10^{-5}	2×10^{-5}

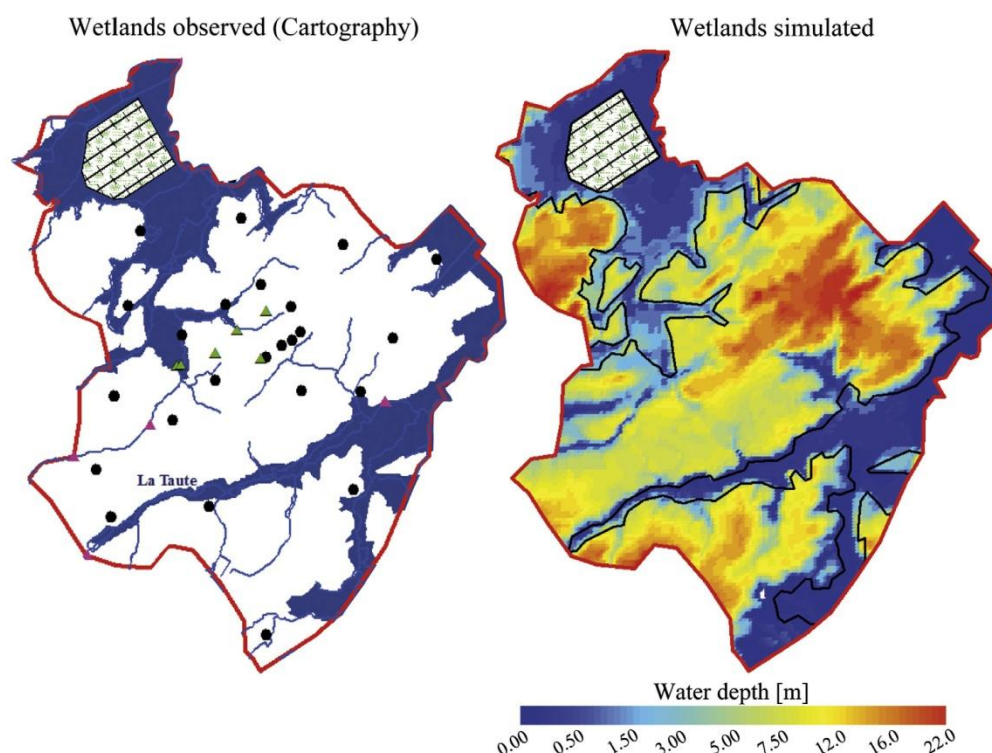


Fig. 6. (A) Observed mean wetlands area. (B) Simulated water table depth over the catchment and related limits of the wetlands area (hydrologic year 2006–2007).

Table 2

Summary of climate change, groundwater abstraction and management scenarios considered in this study. Scenarios are numbered from 1 to 62. The 'Ref' scenario corresponds to no climate change and no groundwater abstraction increase. The letters 'A', 'H' and 'N' for the time slice 2081–2100 correspond to 3 specific GCMs described in Table 3.

Increase of groundwater abstraction (%)	Wells supporting the groundwater abstraction increase	Climatic time slice and GCM				Baupite pumpings
		1961–2000	2081–2100			
			A	H	N	
0	Existing	Ref	9	10	11	ON
10	Existing	1	12	13	14	ON
20	Existing	2	15	16	17	ON
50	Existing	3	18	19	20	ON
100	Existing	4	21	22	23	ON
10	New	5	24	25	26	ON
20	New	6	27	28	29	ON
50	New	7	30	31	32	ON
100	New	8	33	34	35	ON
0	Existing		36	37	38	OFF
10	Existing		39	40	41	OFF
20	Existing		42	43	44	OFF
50	Existing		45	46	47	OFF
100	Existing		48	49	50	OFF
10	New		51	52	53	OFF
20	New		54	55	56	OFF
50	New		57	58	59	OFF
100	New		60	61	62	OFF

3.7. Climate change

Climate change impact studies cannot directly use the output of global climate models (GCM) because of discrepancies between the extent of the impact model (135 km²) and the horizontal resolution of these numerical models (~250 km, see Solomon et al. (2007)). Downscaling methodologies are commonly used to overcome this problem. Such methods are either dynamically and/or statistically based. In the current study, DSCLIM, a statistical downscaling methodology developed at CERFACS was applied to work at an 8 km resolution. This method is based on the physical relation-

ship between large-scale atmospheric circulations and the local-scale climate (Boé et al., 2009; Fowler et al., 2007b; Pagé et al., 2009). Although downscaling methods are also a source of uncertainty (Fowler et al., 2007a), we chose to focus on climate model uncertainty, which has been shown to be conservative for hydrological impact studies in the French context (Ducharne et al., 2009). This was applied to produce climate scenarios for specific chosen locations in France. Using this methodology, 14 GCMs were downscaled from the 2007 CMIP3 database for the A1B greenhouse gas (GHG) scenario over the periods 1961–2000 and 2081–2100, as shown in Table 3 (see Solomon et al., 2007, for details about the

GCMs). Scenario A1B is a sub-category of storyline A1. The A1B scenario was selected because of the availability of several A1B downscaled scenarios, which make possible the evaluation of uncertainties.

In the framework of this study, precipitation and PET time series for 64 downscaled cells corresponding to the Cotentin region, for the 14 GCMs, and the periods 1961–2000 and 2081–2100, were extracted. The period 1961–2000 is considered as the reference case (Table 3). For each cell, each climatic model and each period, the mean annual recharge is calculated using the calibrated GR2M module. The mean recharge rate for the modeled area is then calculated as the average of the 64 cell results. Fig. 7 shows temperature and precipitation changes for the period 2081–2100. Compared to the reference period, the mean annual temperature is expected to increase by between 1.3 and 3.7 °C, and annual precipitation is expected to decrease by between 1.8% and 21.3%. The calculated recharge for the reference case is 250 mm/yr. All climatic projections induce a decrease in recharge rate ranging from 22% to 61%, with an average of 40% (Table 3). Scenarios 'csri_mk3_0' and 'ipsl_cm4' give the minimum and maximum decreases in recharge, respectively.

3.8. Coupling climate change, groundwater abstraction and management

In this impact study, the 3 most contrasted climatic projections, from the 14 downscaled GCMs, are used as input for the hydrological model to investigate the variability between the climate change models (Table 3):

- The most favorable ('csri_mk3_0', termed scenario A, $R = 194$ mm/year).
- The most unfavorable ('ipsl_cm4', termed scenario N, $R = 97$ mm/year).
- The average scenario ('miroc3_2_medres', termed scenario H, $R = 148$ mm/year).

Each climate scenario is coupled with 4 groundwater extraction scenarios (increase of 10%, 20%, 50% and 100%), according to the actual trend in water demand and consumption. Each of these combinations is applied to the two abstraction-management schemes described above: pumping increase in Sainteny (scenarios 12 to 23) or Marchésieux (scenarios 24–35) sub-catchments.

One of the main objectives was to provide insights into wetland management solutions to mitigate climate change and anthropic

impacts. Exploitation of the Bauppte peatland (Fig. 1) stopped in 2006 but pumping is maintained to avoid flooding the surrounding fields which are used for agriculture. An efficient management of Bauppte might therefore provide a solution to reduce negative anthropogenic impacts i.e. water drawdown. The feasibility of this management scheme was studied by complementing the previous scenarios with additional scenarios where pumping is stopped in the Bauppte peat exploitation. This stop is simulated in the model by removing the prescribed head boundary condition over the 0.4 km² 'Bauppte' area. As in the previous cases, the simulation was applied to the two abstraction-management schemes i.e. increased pumping in the Sainteny (scenarios 39–50) or Marchésieux (scenarios 51–62) sub-catchments. Three additional scenarios were tested to estimate the respective impacts of past climate change (increase of 1 °C between 1950 and 2012) and past anthropogenic activities (Pumping of Bauppte and groundwater abstraction) in relation to the current situation.

4. Results

The wetlands compartment corresponds to a natural aquifer outflow and, as shown in the next section, any change in the aquifer recharge or groundwater abstraction is likely to affect wetland area. Future scenarios for groundwater abstraction, climate change and management were then applied as model input. All results for future scenarios were compared to the reference model (1961–2000) with particular focus on the wetland surface area and on water level changes. A summary of these results is presented in Figs. 8 and 9. Proportions of the different water balance terms for some scenarios are given in Table 4.

4.1. Wetland surface reduction

Fig. 8 shows the proportion of wetland surface area for the reference model (1961–2000), for future groundwater abstraction scenarios, climate change scenarios (2081–2100) and coupled scenarios. For the reference model, the proportion of wetland area is equal to 24.4% of the total area. This proportion clearly decreases with increasing groundwater abstraction. If these new groundwater volumes are pumped in existing wells, the wetland area decrease ranges from –0.02% to –3.7% (0.03 km² and 5.05 km²), according to the magnitude of groundwater abstraction. Conversely, if additional pumping is carried out in new wells in the Marchésieux sub-basin, the impact on wetland area is less important and ranges from –0.04% to –1.56% (0.05 km² and 2.1 km²). It is partly due to the better distribution of abstracted volumes over the whole area but also because of the groundwater fluxes entering through the southwest catchment limits which feed the aquifer.

Simulations of the impacts of climate change indicate a significant reduction of wetland surface area by the end of the century (2081–2100), which is correlated to the decrease of recharge (ranging from –22% to –61%, see Table 3). Considering unchanged groundwater abstraction, reduction of the wetland surface area ranges from –13.64% (18.4 km²) for the worst climatic scenario N (recharge of 97 mm) to –5.34% (7.2 km²) for the most favorable scenario A (recharge of 194 mm). These results also provide important information about the respective influence of groundwater abstraction and future climate change. On the scale of the modeled area as a whole, climate change generally induces a larger reduction in wetland area, than any of the groundwater abstraction scenarios. Although scenario A is the most "favorable" climatic scenario, the impact on wetland surface area is actually more important than the worst groundwater abstraction scenario. As shown in Fig. 8, the combined impact of climate change and groundwater abstraction is even more important and ranges from

Table 3
GCMs used for climate projections, related recharge and percentage of decrease relative to current recharge. Climate scenarios A, H and N correspond to the mean and extreme scenarios regarding recharge results.

Scenario	Scenario name	Calculated recharge (mm)	Calculated recharge decrease by 2081–2100 (%)
Ref.	Reference	250	0
A	csri_mk3_0	194	22
B	mri_cgcm3_2a	182	27
C	giss_model_e_r	182	27
D	ccma_cgcm3_1_t63	171	32
E	ingv_echam4	171	32
F	mpi_echam5	171	32
G	giss_aom	163	35
H	miroc3_2_medres	148	41
I	ncar_ccsm3_0	143	43
J	gfdl_cm2_1	137	45
K	gfdl_cm2_0	118	53
L	cnrm_cm3	110	56
M	miub_echo_g	108	57
N	ipsl_cm4	97	61

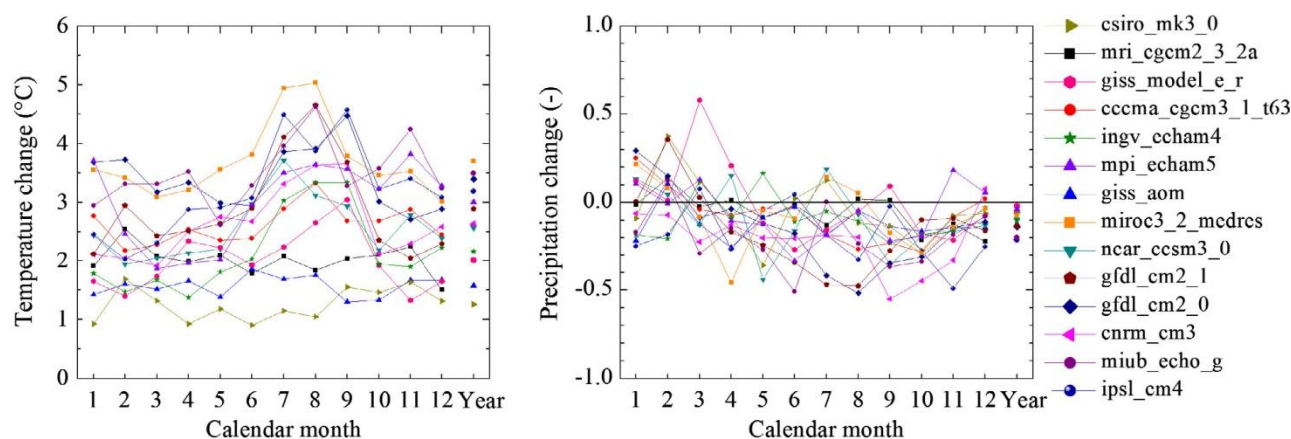


Fig. 7. Monthly and annual mean temperature and precipitation changes for the 14 climatic models in the Cotentin area. Calendar months are numbered from January to December.

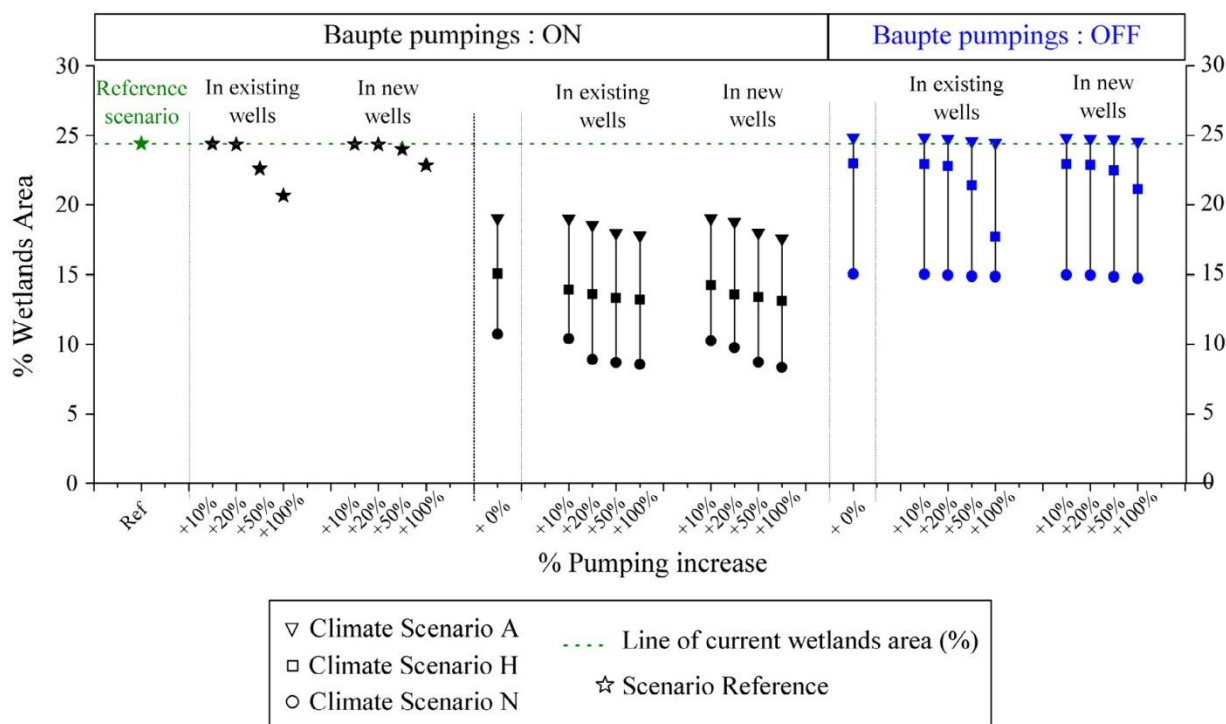


Fig. 8. Percentage of calculated wetlands area in the modeled zone, according to scenarios of groundwater abstraction, climate change and management.

–5.36% to –16.04%, depending on the climate change scenario and location of the pumping wells.

Regarding water balance terms, fluxes entering the domain correspond to the recharge applied on the top cells and groundwater entering by specified flux boundary condition (See Section 3.1). These specified groundwater fluxes represent 36% of total water influx in the modeled domain. Fluxes leaving the domain correspond to the groundwater discharge, pumpings in the public water distribution wells and pumpings in the Baupte peat exploitation. For the reference scenario, these terms correspond to 72%, 9% and 19% of total influx, respectively. Numerical simulations allow quantifying the absolute and relative evolution of these terms considering various stresses (Table 4). For climate change scenarios with unchanged groundwater abstraction, absolute values of all terms logically decrease with recharge and more extreme climate change. However, the proportion of abstracted groundwater (public wells and Baupte) relatively to total influx increases to the detriment of groundwater discharge. For the worst climate change scenario

(scenario N), groundwater discharge is only 32.1% of total influx. In cubic meters, this represents a decrease of more than 80% compared to the reference simulation. Finally, Table 4 also shows that the decrease of total water influx in climate change scenarios is greater than groundwater abstraction by public wells, which partly explains the preponderant impact of climate change on wetland areas. Relations between input stresses and hydrogeology variables are however complex and dependent on many parameters (such as geology, topography, locations of pumping wells), so that numerical modeling is required for an objective impact quantification.

4.2. Wetland spatial distribution

Previous analyzes provide overall information on the scale of the modeled area. However, the different scenarios also imply different impacts in terms of the distribution of drawdown within the wetlands (Fig. 9).

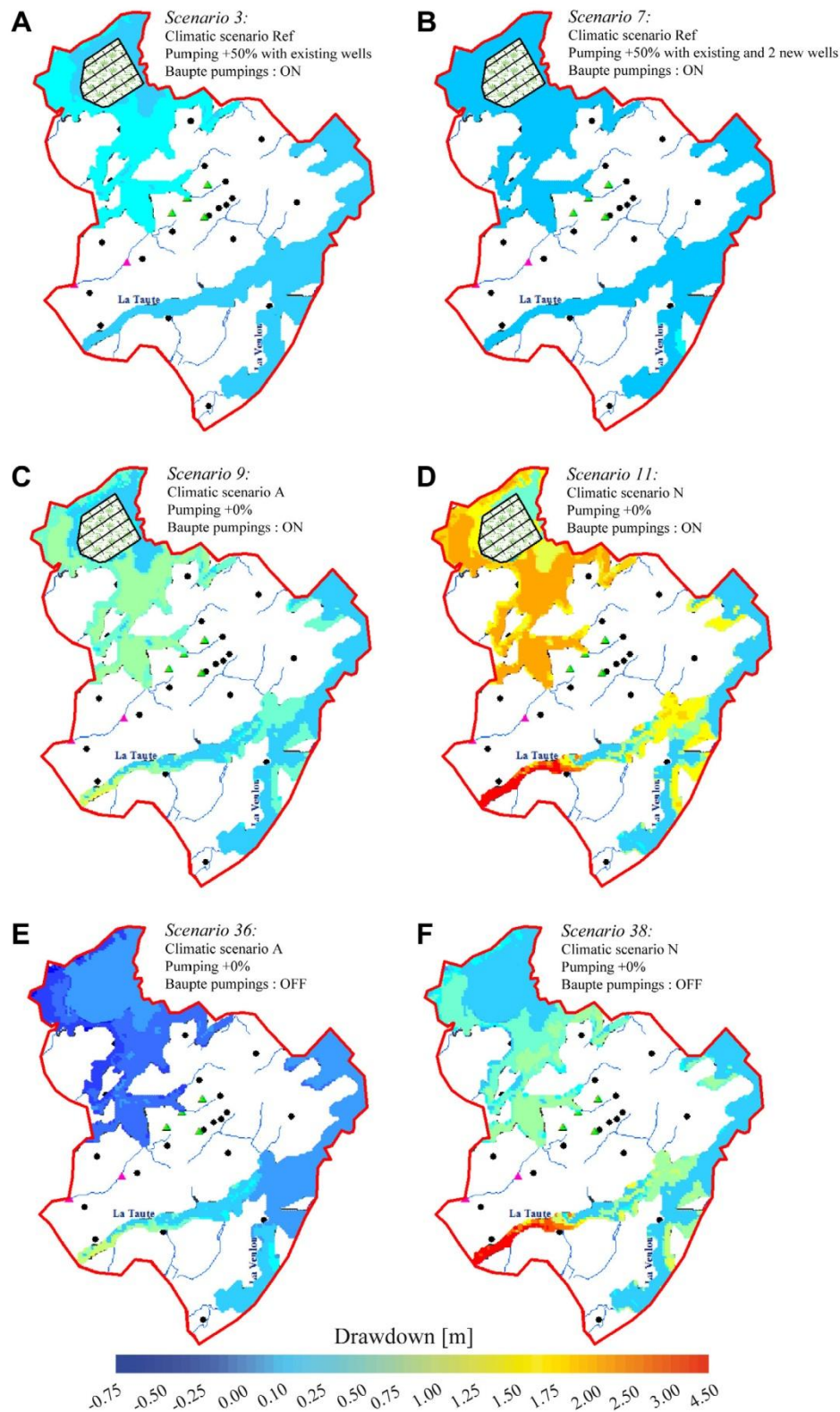


Fig. 9. Maps of drawdown for different climate change and management scenarios.

Increasing pumping rates by 50% in existing wells induces a water level decrease of between 25 cm and 40 cm in the Sainteny Northern wetland, while water levels are not significantly affected in the Marchésieux Southern wetland (Fig. 9A, Scenario Pumping +50% with existing wells, Scenario 3). Conversely, increased pumping in new wells located in the Marchésieux basin leads to a better distribution of the impacts (Fig. 9B, Scenario Pumping +50% with

existing and 2 new wells, Scenario 7). The impact of climate scenarios is greater but better distributed over the whole area (Fig. 9C, Scenario A and Fig. 9D, Scenario N). For climate change scenario A, water levels in the Northern wetland are lowered by about 50–60 cm. In the Southern area, the water levels decrease by 25–40 cm in the east and by 80–90 cm in the most elevated part of the modeled area. Generally, wetland areas are more impacted

Table 4

Main water balance terms for the reference and climate change scenarios.

Climate Scenario		Total influx	Groundwater discharge	Public wells	Bauppte
Reference (R = 250 mm/yr)	m ³ /yr	5.2E + 07	−3.7E + 07	−4.9E + 06	−1.0E + 07
		100	−71.3	−9.4	−19.2
A (R = 194 mm/yr)	% Of total influx	75	−46.9	−9.4	−18.7
H (R = 148 mm/yr)	% Of reference total influx	56	−28.7	−9.4	−17.7
N (R = 97 mm/yr)		38	−12.1	−9.4	−16.2
A (R = 194 mm/yr)	% Of total influx	100	−62.6	−12.6	−24.9
H (R = 148 mm/yr)		100	−51.4	−16.9	−31.7
N (R = 97 mm/yr)		100	−32.1	−25.0	−42.9

in the Northern catchment, where groundwater levels are also affected by the Bauppte peat exploitation. In this catchment, all flooded areas disappear with the worst climate change scenario (scenario N).

These results show the possible evolution of the wetland area according to different groundwater abstraction options and climate change scenarios for the end of the century (2081–2100). The wetland area is expected to decrease in any case, and the impact of climate change is stronger than the impact of groundwater abstraction.

4.3. Effect of pumping in the Bauppte peat exploitation

One potential solution to save water and mitigate climate and pumping impacts would be to reduce pumping in the Bauppte peat exploitation (Fig. 8). For all scenarios, stopping all pumping in Bauppte would allow a wetland recovery of 4.45–9.19% of the total area depending on the groundwater abstraction and climate change scenario. Considering the most favorable climate change scenario (Scenario A), the wetland surface area would be approximately equivalent to the current situation, whatever the pumping scenario used (see Fig. 8). This effect is apparent in Fig. 9E, Scenario 36, where water levels increase by 75–25 cm in the Sainteny area. For the other scenarios, stopping all pumping in Bauppte is not enough to completely balance the loss of wetlands due to climate change. Considering the worst scenario (climate scenario N and Pumping + 100% with existing wells), reduction of the wetlands area would still attain 9.67%, even if pumping in Bauppte is stopped (Fig. 9F).

5. Discussion

5.1. Uncertainty and model limitations

Using numerical models induces some uncertainty that affect the subsequent simulations. This uncertainty may be generated from various possible sources (Refsgaard et al., 2006). Some of them are discussed here below. By adopting a multi-model approach for the climate scenarios, it is possible to incorporate the uncertainty related to the climate models and the uncertainty derived from climate model selection into the assessment of climate change impacts on the Sainteny-Marchésieux catchment. All the 14 climate change scenarios predict a decrease in recharge ranging from 22% to 61% (Table 3). It results in a decrease of water level and total wetland surface area ranging from 5.3% to 13.6% (Fig. 8), meaning that this decrease is highly probable from this point of view.

The accuracy of the predictions will also depends on the quality of the calibration, which varies according to the different variables considered in the study. The volume of drained water presents the major uncertainty because only partial observed stream-discharge data are available. In spite of the lack of data, the 3D hydrogeological model satisfactorily reproduces the measured volumes of

drained water with a good correlation ($R^2 = 0.9$) between simulated and observed values (error less than 5%). Similarly, the volumes leaving the system by the Bauppte boundary condition match the measured quantity of water currently pumped from the peat exploitation. Concerning the hydraulic heads (Fig. 5), all residuals are lower than 1 m, except for two wells. The model is able to simulate groundwater levels according to different annual climate conditions, even though it slightly over-estimates the hydraulic heads in wet periods and under-estimates them in dry periods, which could also imply that the predicted impacts are slightly overestimated.

We here emphasize the relative simplicity of the model, which is focused on the evolution of wetlands extension. Particularly, the use of a seepage boundary condition for the whole modeled surface enables some flexibility regarding the distribution of discharge zones over the domain. These discharge zones are actually variable according to climatic conditions. As an example, low recharge rates induce lower water table and disconnection of river sections, which also implies a decrease of groundwater discharging zones and wetlands areas (Goderniaux et al., 2013). Conceptualizing and representing these processes in the numerical model is crucial to quantify the extents of these wetland areas as a function of recharge. Specifying the locations of rivers and using river boundary conditions appears too restrictive in this case. Although simple, the approach adopted however provides a rapid and easy characterization of wetland extension, which is clear and important parameter for stakeholders.

More complex approaches are available for modeling hydrological systems. Integrating more processes into the same model has the advantage of providing more realistic simulations. Indeed it is very useful to have realistic water budget terms. That's why, particularly, fully integrated surface–subsurface models are more and more used (Ebel et al., 2009; Jones et al., 2008; Liggett et al., 2013). However, using more complex hydrological models also involves a large number of parameters, requires important computing times, and makes the calibration step more difficult, so that significant uncertainty may remain from this source. There is a lively debate on the question of the models complexity to be used (Hill, 2006; Hunt and Zheng, 1999). In this study, the model used includes simplifications, which presents some advantages but also some limitations. The processes related to the water transfers in the partially saturated zone are for example not simulated by the hydrological model. The role of these transfers is limited regarding the results of this study because simulations are performed in steady state and the partially saturated zone remains relatively thin. However, for transient simulations, and particularly to evaluate seasonal fluctuations, water transfers in this zone should be further studied. Similarly, the verification of the water budget terms for the GR2M recharge model and the Modflow hydrological model is currently based on annual data. A finer time-discretization would be required to account for seasonality effects. Moreover, more observed data about wetlands extents at the seasonal timescale would also be required. While this study has shown the long term effect of climate change on wetland areas, the implications regarding these

seasonal fluctuations remain to be studied and constitute a perspective of this work.

5.2. Groundwater abstraction and climate change scenarios

The groundwater abstraction scenarios were implemented to evaluate the sensitivity of the Cotentin wetlands to future increasing demand. The pumping simulations reflect realistic scenarios of future exploitation, according to local water agencies. In general, pumping in the main aquifer decreases upward fluxes (from the aquifer to the peat) and increases downward fluxes (from the peat to the aquifer). These modifications of water transfer from one compartment to the other may affect water and peat chemistry. Enhanced downward fluxes will actually bring different water, with higher oxygen content and different composition, to the deeper peat layers which may, in turn, affect peat structure, mineralization processes, and water quality. Pumping scenarios which include new extraction wells in the Marchésieux sub-catchment should therefore be preferred to limit environmental impact (see Fig. 8 and Fig. 9). Although this hydrological basin is less permeable, a similar water volume abstracted (relative to the amount currently extracted at Sainteny) results in a smaller reduction of the wetland water level (Fig. 8). Moreover, future increased exploitation should remain below a threshold of 10–20% of the current extracted volume to limit the potential impact on wetland surface area.

The 14 climate change scenarios predict a decrease in recharge ranging from 22% to 61% (Table 3) which results in a decrease of total wetland surface area of 5.3–13.6% (Fig. 8). In the long term, the model results clearly show and quantify that the water stresses and the impact on the wetland extents are much greater for the climate change scenarios than for the groundwater abstraction scenarios.

Therefore, climate change constitutes a major driver as compared to groundwater exploitation in the modeled area. However, the effects of climate change will be gradually visible over several decades, whereas the other effects are already severe. Furthermore, as all anthropogenic effects are cumulative, the expected impacts of climate change should emphasize the urgent need for mitigation plans. In this context, the modeling results also highlight the effect of the Baupré exploitation on peat water levels. Peat extraction was stopped in 2006. However, local authorities decided to maintain water pumping in order to avoid flooding agricultural fields. In the near future, pumping could be decreased in order to mitigate the impacts of climate change in the Northern Sainteny catchment.

Thompson et al. (2009) found similar conclusions regarding climate change impact on a wetland area located in south-eastern England, with significant wetland area decrease by the 2050s. The comparison is however difficult as the influence of the groundwater compartment seems less preponderant in their study area. Other studies do not directly calculate wetland extents, but concentrate on groundwater levels and discharge rates evolution. Candela et al. (2009) project decreases in spring discharges to a wetland in Majorca (Spain), for 2025 and 2 emission scenarios (A2 and B2). They calculate that a reduction or alternative management of the groundwater abstraction is needed to avoid the partial or complete disappearance of the wetland. Finally, Herrera-Pantoja et al. (2012) calculated significant declining trends in groundwater levels in a wetland located in Eastern England, by the end of the century and using a 'high' greenhouse gases emissions.

5.3. Anthropogenic influences prior to 2012

During the last years, it was often claimed that groundwater extraction was responsible for peatland desiccation and geotechnical damages. To provide a scientific basis to this controversy, the model has been used to analyze the respective effects of anthropo-

genic activities on groundwater levels over the period 1950–2012. The effects of both the Baupré peatland exploitation and groundwater abstraction were analyzed by removing both pumping from the current situation. The effect of climate change was considered by assuming an increase in annual temperature of 1 °C from 1950 to 2012, as observed on several climatic stations in the region. The recharge and hydrological models were run with a temperature one degree lower than the current temperature. The results indicated a general decrease in water level, in the investigated zone, between 1950 and 2012. Baupré exploitation and groundwater abstraction had relatively similar impacts ranging from 50 to 85 cm and 35 to 70 cm, respectively. Climate change had a more limited impact of about 20 cm over the last 60 years.

The model developed in this study provides interesting insights in the quest to find solutions for this territorial management crisis. It enables the respective impacts of all human activities for the last 60 years to be quantified. The decrease in water-level was reported by local inhabitants, but its extent and the period of occurrence remained unclear. Although the effect of drainage which occurred from the 17th century onwards and more intensively after the Second World War, could not be taken into account, the model results show that more recent human-induced changes have in any case had a major effect during the last decades independently of previous management schemes. Clearly, none of the three anthropogenic effects considered (Baupré exploitation, groundwater exploitation, and climate change) can alone be considered as responsible for peat desiccation. The current state of the peatland appears to result from increasing stress which has several causes. The model results were particularly unexpected for the end-users, who had mainly focused on the impact of groundwater exploitation and had never integrated the potential influence of climate change. This result is particularly important with regard to previous studies which had already indicated severe drawdown (Auterives et al., 2011) and chemical oxidation of the peat (Bougon et al., 2011; De Ridder et al., 2012).

6. Conclusion

The water fluxes occurring between large wetlands and underlying aquifers were analyzed by modeling. A simple model was used to simulate groundwater levels, river fluxes through the wetlands and wetland surface extension. The surface flooded is an important parameter for wetland management and special emphasis was given to this variable. It was computed by applying the seepage boundary condition to the entire area modeled, and measuring the water level in the wetland aquifer.

The model was used to analyze three different anthropogenic effects: (1) groundwater exploitation in the underlying aquifer, (2) wetland water abstraction in a peat exploitation quarry, and (3) the impact of climate change using data from 14 downscaled climate models.

A 100% increase in the groundwater abstraction rate had a maximum impact of 3.7% on the current wetland surface. Climate change is expected to have a greater impact with potential reduction of the wetland surface area ranging from 5.34% to 13.64%. Although peat exploitation has ceased, water pumping has been maintained to avoid flooding farmland. The model indicates that the climate change effects could be partly compensated by decreasing and then stopping this pumping.

Finally, in order to understand the origin of the geotechnical damage observed in recent years, the model was used to investigate the respective impacts of different anthropogenic activities prior to 2012. Results revealed that during the last 60 years, a wetland water-level decrease of 40–90 cm could be attributed to the combined impacts of groundwater and peatland water

exploitation. It is clearly apparent that all these human activities contribute to lower the peat groundwater level and have already severely destabilized peat functioning. All these activities have to be taken into account in future management strategies which it is urgent to define. Water management will require an appropriate compromise which encompasses ecosystem preservation, economic and public domain activities.

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3.3. Conclusion

Cette étude a permis de caractériser les interactions se faisant entre une grande étendue de zones humides et la ressource en eau souterraine de l'aquifère sous-jacent. La forte dépendance de ces compartiments du système hydrologique est mise en évidence via une modélisation numérique 3D des écoulements permettant l'analyse des flux d'échanges entre l'aquifère et les zones humides. Ce modèle numérique est utilisé pour simuler les niveaux d'eau dans l'aquifère, les flux transitant par les zones humides (avec comme exutoire les rivières) ainsi que l'étendue des surfaces inondées.

Les résultats produits, basés sur une analyse de l'étendue des zones humides permettent de mettre en évidence l'impact respectif (passées et futures) des variations climatiques, de l'exploitation de la ressource en eau souterraine et des pompages de la tourbière. Plus particulièrement, la prédominance de l'impact du climat à venir en comparaison avec l'augmentation supposée de la demande en eau souterraine est mise en évidence. En effet, l'impact dû à un doublement des volumes pompés actuels résulterait en une perte de plus de 3.5 % de la surface totale des zones humides actuelles alors que le changement climatique pourrait réduire leurs surfaces de 5 à plus de 13.5 %.

D'autre part, la modélisation permet de proposer des solutions de gestion afin de limiter la perte de zones humides, notamment en jouant sur les pompages au sein de la tourbière. Il semble même que dans le cas d'un impact climatique plutôt « modéré » l'arrêt des pompages au sein de la tourbière permettrait de maintenir la surface actuelle de zones humides.

Enfin, les résultats des simulations numériques permettent la mise en relation de l'affaissement des sols observés avec la réduction des niveaux d'eau au sein des zones humides sur les dernières décennies (Figure 31), dont la responsabilité pourrait être attribuée aux pompages (dans l'aquifère et dans la tourbière) et donc plus globalement aux activités anthropiques.



Figure 31. Affaissement de sol marqué par l'écart entre le sol et la base du poteau électrique (source : <http://www.cc-carentan-cotentin.fr>).

En conclusion, cette étude montre d'une part l'importance des activités humaines sur les stocks d'eau et donc sur le fonctionnement global du système hydrogéologique. D'autre part, il convient de remarquer que l'impact du climat reste un facteur primordial, c'est pourquoi la prise en compte des activités anthropiques et de l'évolution possible du climat sont nécessaires dans tout plan de gestion durable de la ressource en eau.

Chapitre 4. Relation eau souterraine – eau de mer et climat

4.1. Introduction

En plus des interactions avec les divers compartiments de surface (rivières, lacs et zones humides), les eaux souterraines interagissent aussi avec les océans. Les aquifères côtiers constituent l'ultime zone de décharge des systèmes hydrogéologiques régionaux (Porter et al., 2008). Ce processus est caractérisé par deux phénomènes principaux qui sont d'une part la décharge d'eau souterraine vers l'océan et d'autre part l'intrusion d'eau de mer dans l'aquifère. Sous l'effet de la différence de densité entre l'eau de mer et l'eau douce, l'eau de mer (plus dense) pénètre à l'intérieur des terres jusqu'à atteindre un équilibre avec la décharge d'eau souterraine. Ce phénomène se manifeste par la création d'une interface eau douce-eau salée pouvant s'étendre sur plusieurs kilomètres à l'intérieur de l'aquifère (Prieto, 2005) (Figure 32).

La première approximation physique de ce phénomène fut établie par Ghyben-Herzberg (Ghyben, 1889; Herzberg, 1901), donnant naissance à une solution analytique permettant d'estimer la profondeur de l'interface (eau douce-eau de mer).

$$z_s = \frac{\rho_0}{\rho_s - \rho_0} z_w$$

avec z_s la profondeur de l'interface par rapport au niveau de la mer, z_w l'altitude du niveau de la nappe au-dessus du niveau de la mer, ρ_s la densité de l'eau de mer et ρ_0 la densité de l'eau douce (Figure 32). Considérant, la densité de l'eau de mer $\rho_s = 1025 \text{ kg.m}^{-3}$ et celle de l'eau douce $\rho_0 = 1000 \text{ kg.m}^{-3}$ donne :

$$z_s = 40. z_w$$

Néanmoins, cette relation n'est valable que dans le cas d'un aquifère libre homogène et ne prend pas compte les nombreuses perturbations naturelles (comme les marées) et anthropiques (pompages) modifiant la position et les caractéristiques de l'interface.

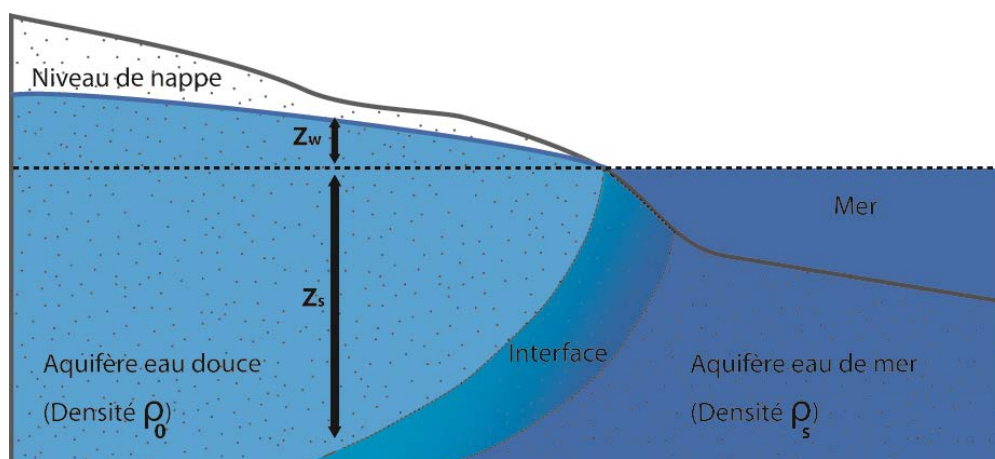


Figure 32. Schéma descriptif de la relation entre l'eau de mer et les eaux souterraines.

Ces premiers travaux ont depuis donné naissance à de nombreuses études du fait du grand intérêt de la ressource en eau des aquifères côtiers, permettant de bien comprendre la nature de ces interactions (Bear et al., 1999a). En outre, elles ont permis de mettre en lumière que les processus de salinisation des eaux souterraines sont gouvernés par la différence de densité entre l'eau douce et l'eau de mer et par leur distribution relative au sein du système hydrogéologique. Fonction de cette distribution, l'intrusion d'eau de mer peut se produire selon différents mouvements principalement caractérisés par :

- mouvement latéral de l'eau de mer vers l'intérieur des terres au sein de l'aquifère, équivalent à l'avancée du biseau salé.
- infiltration verticale descendante du fait de la mise en place et de la présence d'eau de mer au-dessus d'un aquifère d'eau douce, par exemple transgression marine.
- mouvement vertical ascendant engendrant la remontée d'une partie souterraine avec de l'eau plus salée, comme la remontée du biseau salé du fait de pompage dans l'aquifère côtier.

L'intrusion d'eau de mer est d'une part engendrée par des perturbations de niveau d'eau au sein des aquifères sur des périodes prolongées et d'autre part par des événements épisodiques (Werner et al., 2013). Les processus naturels comme les mouvements tectoniques et les variations climatiques se déroulant sur des temps géologiques (caractérisés par des variations eustatiques) ont entraîné la salinisation d'aquifères (Carretero et al., 2013; Chang et al., 2011; Gossel et al., 2010; Maas, 2007;

Sherif and Singh, 1999; Watson et al., 2010; Webb and Howard, 2011; Werner and Simmons, 2009). Sur une autre échelle de temps, les événements naturels épisodiques comme les tsunamis (Illangasekare et al., 2006; Violette et al., 2009), les tempêtes (Terry and Falkland, 2009; Wilson et al., 2011) ou périodiques comme les marées (Ataie-Ashtiani et al., 1999; Werner and Lockington, 2006) ont été identifiés comme étant responsable de salinisation. Enfin, les activités anthropiques comme la récupération des aires côtières pour diverses activités (agriculture, industrie...), ainsi que le pompage d'eau souterraine perturbent les conditions hydrologiques du système entraînant la redistribution des eaux douces et salées (Calvache and Pulido-Bosch, 1997; Jakovovic et al., 2011; Luyun et al., 2011; Pool and Carrera, 2011; Reilly and Goodman, 1987).

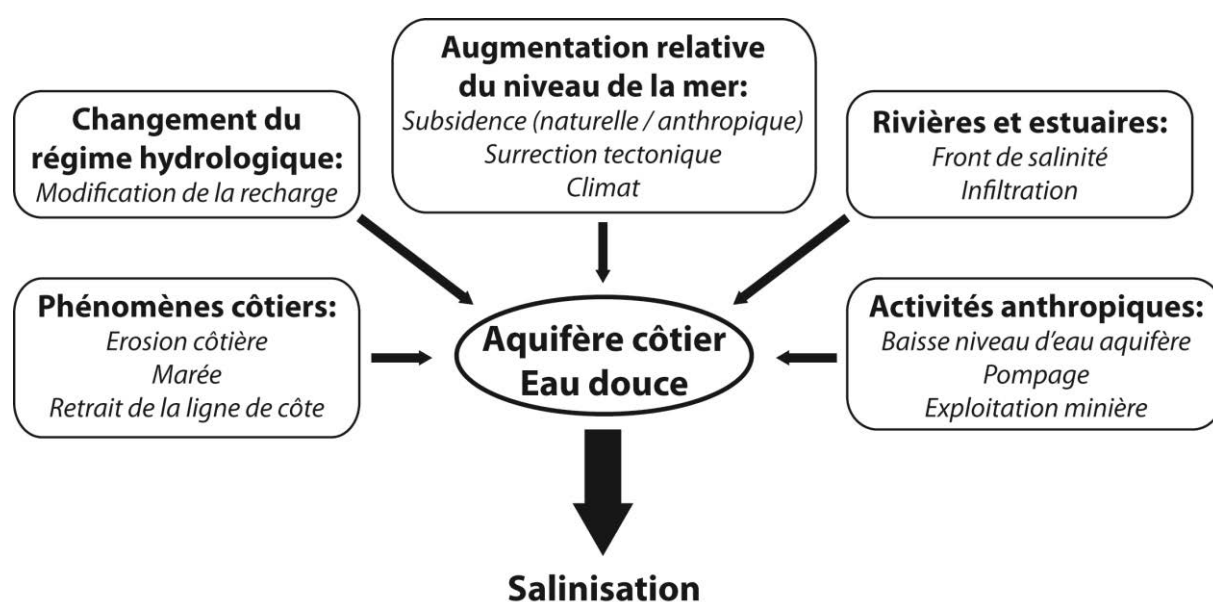


Figure 33. Processus naturels et anthropiques pouvant être responsables de la salinisation des aquifères, modifiée à partir de Oude Essink (2001).

La salinisation affecte les aquifères côtiers du fait de leur relation particulière avec l'océan et peut être la conséquence de divers processus aussi bien naturels, qu'anthropiques (Custodio et al., 1987; Oude Essink, 2001) (Figure 33). La combinaison des divers processus naturels avec les nombreuses activités anthropiques génère un nombre presque infini de situations possibles pouvant entraîner la salinisation des aquifères (Werner et al., 2013).

En conclusion, il est à remarquer que l'étude des processus de salinisation s'est focalisée sur les aquifères côtiers et les divers processus pouvant entraîner la modification de l'équilibre hydrologique existant. Comme souligné par Werner et al. en

2013 la salinisation des compartiments d'eau de surface du fait de la pénétration de l'eau de mer dans ceux-ci (progression du front de salinité) pourrait significativement contribuer à la salinisation des aquifères. Or, l'étude de la salinisation de ces compartiments, comme les rivières ou fleuves, n'a actuellement pas été prise en compte. Le travail proposé qui suit s'inscrit dans cette démarche visant à considérer les compartiments d'eau de surface et plus particulièrement le système fluvial dans les possibles mécanismes de salinisation des aquifères.

4.2. Modélisation numérique des écoulements à densité variable avec COMSOL Multiphysics

L'étude des processus de salinisation s'appuie principalement sur la modélisation numérique et les solutions analytiques ainsi que sur les études géochimiques. Les modèles d'intrusion d'eau de mer (modélisation à densité variable) permettent de prendre en compte les variations de densité. Malgré une différence de densité de seulement 2.5% entre l'eau de mer et l'eau douce cette variation constitue le point clé de ces problématiques. La modélisation des écoulements à densité variable est donc le point fort de l'approche numérique, contrairement aux solutions analytiques qui ne sont basées que sur l'hypothèse d'interface eau de mer/eau douce. De plus, fait du nombre limité de solutions analytiques, la modélisation à densité variable reste l'outil indispensable et le plus efficace pour l'étude des processus de salinisation.

De nombreux problèmes environnementaux requièrent la prise en compte de la dynamique des eaux saumâtres et de l'eau de mer comme le stockage de déchets (toxiques et radioactifs) dans les milieux fracturés ou dans les formations de roches salées, le lessivage de déchets industriels et l'intrusion d'eau marine dans les aquifères côtiers ou les aquifères surplombés par des formations salines. C'est pourquoi depuis un certain nombre d'années de nombreux codes ont été développés pour simuler les écoulements des eaux souterraines à densité variable (Kolditz et al., 1998). Les codes numériques peuvent être vérifiés par comparaison avec les solutions analytiques et par la méthode du « benchmarking ». Cette méthode consiste à comparer les sorties du modèle numérique avec des problèmes standards bien définis (Simpson and Clement, 2004).

Afin d'attester la capacité des modèles à reproduire ces processus physiques et plus particulièrement les écoulements à densité variable, différents benchmarks de référence ont vu le jour. L'étude des cas-tests bien documentée est très utile pour la vérification des modèles numériques (Kolditz et al., 1998). À ce jour, il existe plusieurs cas-tests pour les problèmes à densité variable 2D (Diersch and Kolditz, 2002) : The Henry (Henry, 1964), Elder (Elder, 1967), Salt dome (Konikow et al., 1997, 1996), Salt lake (Simmons et al., 1999) problems.... Néanmoins, « The Henry problem » reste le seul benchmark qui possède une solution analytique pour les écoulements à densité variable.

Pour attester la capacité de COMSOL Multiphysics (logiciel de modélisation à élément fini permettant de résoudre une large gamme d'équations différentielles) à modéliser les écoulements à densité variable, 3 benchmarks ont été utilisés afin de traiter différents aspects des écoulements à densité variable en relation avec les problématiques de cette thèse (instabilité densitaire, biseau salé et lessivage). Les 3 benchmarks et leurs résultats obtenus avec COMSOL Multiphysics sont présentés dans les 3 points suivants :

« The Henry problem » (Henry, 1964)

Ce problème d'intrusion d'eau salée est unique dans son genre puisqu'il dispose d'une solution analytique. Ce cas, d'un aquifère confiné, homogène et isotrope, implique une recharge d'eau douce arrivant par la limite gauche (décharge d'eau douce de l'aquifère vers la mer) alors que la limite droite représente la mer. Ce cas fait donc référence au biseau salé (Holzbecher, 1998).

Conditions limites et initiales du modèle standard

Le problème de Henry est représenté par un aquifère rectangulaire. Les limites horizontales (haut et bas) du modèle sont imperméables et correspondent à des limites à flux nul. Un flux d'eau douce est appliqué à la limite verticale gauche (à $x=0$), tandis que pour la limite droite (à $x=2$) une charge hydraulique constante fonction de la densité d'eau de mer est appliquée (Figure 34).

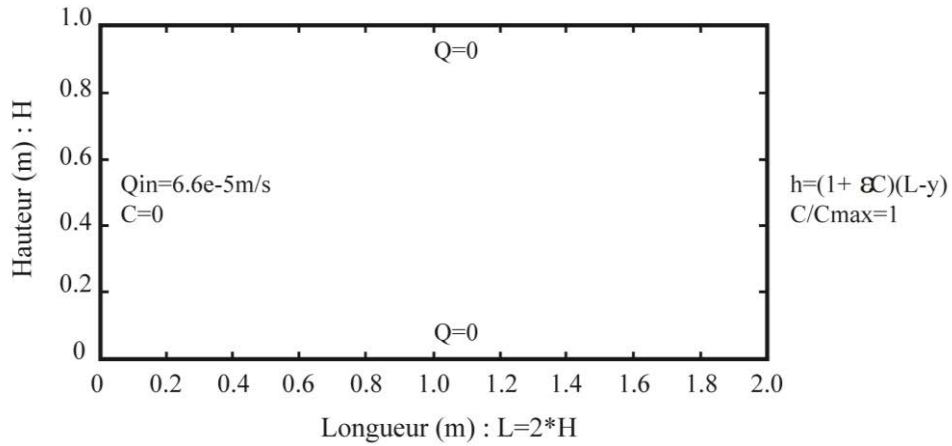


Figure 34. Conditions limites et initiales du problème de Henry.

Au temps $t=0$, le milieu poreux ne contient que de l'eau douce. Le Tableau 2 synthétise les paramètres utilisés pour le problème standard de Henry. Afin de comparer les différents cas de ce problème, des paramètres adimensionnels ont été définis :

$$\begin{aligned} &= \frac{L}{H} \\ a &= \frac{\mu \cdot Q_{in}}{k \cdot g \cdot H \cdot (\rho_s - \rho_0)} \\ b &= \frac{\theta \cdot D_m}{Q_{in}} \end{aligned}$$

avec le ratio des dimensions du domaine avec L la longueur et H la hauteur du domaine, a le paramètre relatif à la recharge avec μ la viscosité dynamique, Q_{in} un apport d'eau latéral, k la perméabilité, g l'accélération de la pesanteur, ρ_s la densité de l'eau de mer et ρ_0 la densité de l'eau douce et b l'inverse du nombre de Péclet avec θ la porosité et D_m le coefficient de diffusion moléculaire. Dans le cas de référence, les valeurs des 3 paramètres sont: $= 2$, $a = 0.264$ et $b = 0.1$ (correspondant aux valeurs du Tableau 2).

Tableau 2. Paramètres de la modélisation pour le problème standard de Henry

Symbole	Paramètre	Valeur	Unité
Dm	Coefficient de diffusion moléculaire	1.886E-05	m ² /s
g	Accélération de la pesanteur	9.81	m/s ²
k	Perméabilité	1.0E-09	m ²
Qin	Recharge	6.6E-05	m/s
θ	Porosité	0.35	
α	Coefficient de dispersivité longitudinale et transversale	0 ; 0	m
0	Densité eau douce (Référence)	1000	kg/m ³
S	Densité eau de mer	1025	kg/m ³
μ	Viscosité dynamique	1.00E-03	kg/m/s
H	Dimension du modèle	1	m

Simulations et résultats

Dans les conditions initiales, l'aquifère ne contient que de l'eau douce ($\rho_0 = 1000 \text{ kg} \cdot \text{m}^{-3}$). Au cours du temps et de l'évolution de la zone saumâtre, l'eau salée entre par la base du côté droit de l'aquifère tout en se mélangeant par diffusion et dispersion avec l'eau douce (effet causé par la différence de densité). Ce mouvement induit par le contraste de densité entraîne la formation d'un écoulement d'eau de mer vers l'intérieur de l'aquifère d'eau douce. D'autre part, l'écoulement d'eau douce en direction de la mer remonte dans la partie haute de l'aquifère (Figure 35). Le mélange causé par la dispersion hydrodynamique (D), limite l'extension de l'eau de mer vers l'intérieur des terres (Cooper et al., 1964).

Du fait de la grande utilité de ce benchmark pour tester les codes, ce cas-test a été le sujet de très nombreuses études dont les résultats ont été synthétisés et mis en comparaison par Soto Meca et al.,(2007). La mise en comparaison des lignes d'isoconcentration simulées par chaque code montre une bonne cohérence des résultats (à la fois entre les résultats numériques et la solution semi-analytique) cependant quelques différences persistent du fait de la technique numérique employée (Soto Meca et al., 2007).

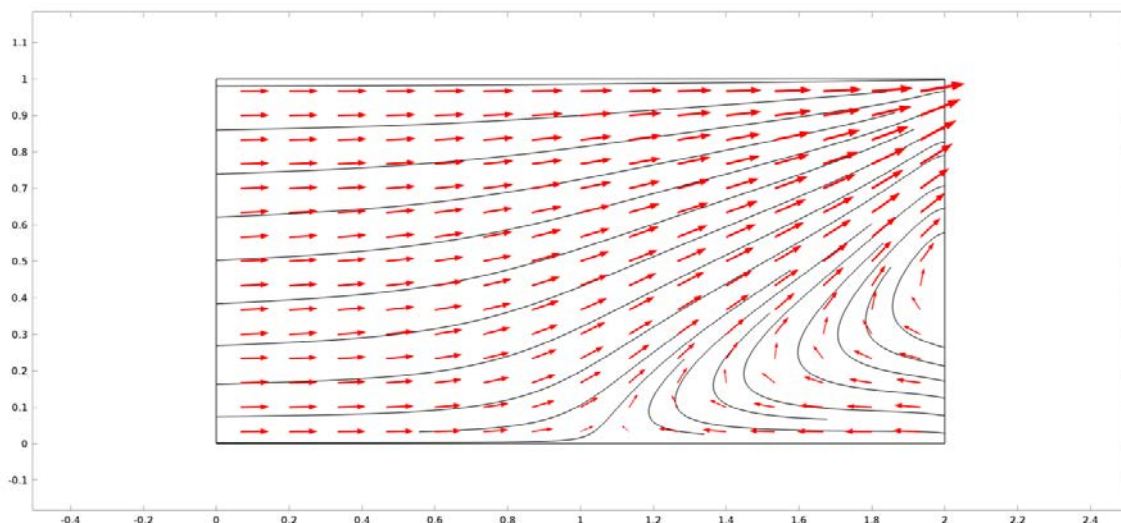


Figure 35. Lignes et direction des écoulements au sein de l'aquifère dans le cas de référence. La taille des flèches témoigne de l'importance de l'écoulement.

Les résultats obtenus avec COMSOL Multiphysics pour le problème de Henry peuvent donc être comparés avec l'ensemble des résultats des précédentes études (Soto Meca et al., 2007). La Figure 36 présente les résultats pour le cas standard ($\xi = 2$, $a = 0.264$ et $b = 0.1$) et un cas modifié du problème de Henry ($\xi = 2$, $a = 0.264$ et $b = 0.2$).

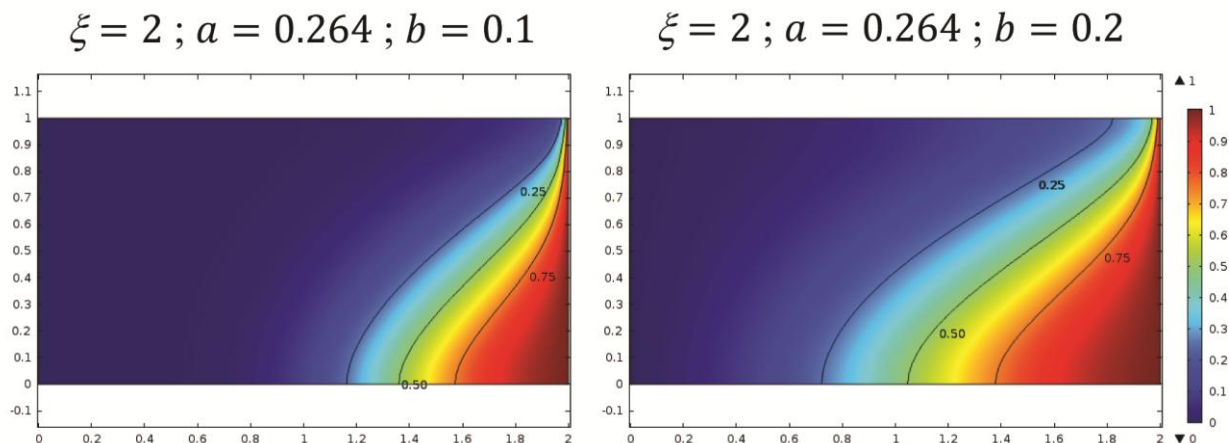


Figure 36. Lignes d'isoconcentration (0.25, 0.5 et 0.75) correspondant à une fraction de la concentration de l'eau de mer. À gauche, le cas standard du problème de Henry (avec $b=0.1$). À droite, le problème de Henry modifié et introduit par Simpson and Clement 2004 (avec $b=0.2$).

La ligne d'isoconcentration 0.5 pour le problème de Henry ($b=0.1$) obtenue avec COMSOL est à la base de l'aquifère à la position $x=1.36\text{m}$ (Figure 36). Cette valeur obtenue est en accord avec les résultats des précédentes études avec des positions comprises entre 1.242m et 1.394m (Croucher and O'sullivan, 1995; Gotovac et al., 2003;

Langevin and Guo, 1999; Lee and Cheng, 1974; Segol, 1994; Simpson and Clement, 2004; Soto Meca et al., 2007).

De même, dans le cas modifié ($b=0.2$), les précédentes études (Langevin and Guo, 1999; Simpson and Clement, 2004; Soto Meca et al., 2007) donnent x entre 1.059 et 1.078m. Or, avec COMSOL nous obtenons $x=1.06$ m.

La cohérence des résultats permet de mettre en avant la capacité de COMSOL à reproduire le problème de Henry, malgré quelques différences ceci étant la conséquence des limites des calculateurs (Segol, 1994). Néanmoins, ce problème seul ne permet pas d'affirmer que le simulateur est totalement capable de gérer les écoulements à densité variable. C'est la raison pour laquelle d'autres cas-tests sont utilisés.

« The Elder problem » (Elder, 1967)

Le problème d'Elder est un problème de convection libre au sein d'un milieu poreux homogène et rectangulaire. À l'origine, ce problème d'écoulement induit par une différence de température a été par la suite adapté pour devenir un problème de transport de soluté à densité variable (Elder, 1967). Ce problème a souvent été utilisé comme benchmark pour la vérification de code, comme SUTRA (Voss and Souza, 1987), FAST-C (Holzbecher, 1998), TOUGH2 (Oldenburg and Pruess, 1995), FEFLOW (Diersch, 1996),etc..

Le problème d'Elder caractérise un processus de circulation initié du fait de la présence d'un soluté plus dense au-dessus d'un moins dense. Le mouvement du soluté au sein du milieu est exclusivement initié par un processus de diffusion (Soto Meca et al., 2007). Cette circulation perdure ensuite sous l'effet de la rééquilibration de la densité permettant ainsi la dispersion du sel au sein de l'aquifère.

Conditions limites et initiales du modèle standard

Le problème d'Elder est représenté par un aquifère rectangulaire clos, signifiant qu'aucun écoulement ne se fait à travers les limites. Uniquement deux points situés dans les coins supérieurs du domaine correspondent à une pression hydrostatique nulle. Une source de soluté de concentration constante est localisée au centre de la limite supérieure du domaine alors que la limite inférieure est maintenue à une valeur de

concentration constante nulle (Figure 37). Au temps $t=0$, le milieu poreux ne contient que de l'eau douce. Le Tableau 3 synthétise les paramètres utilisés.

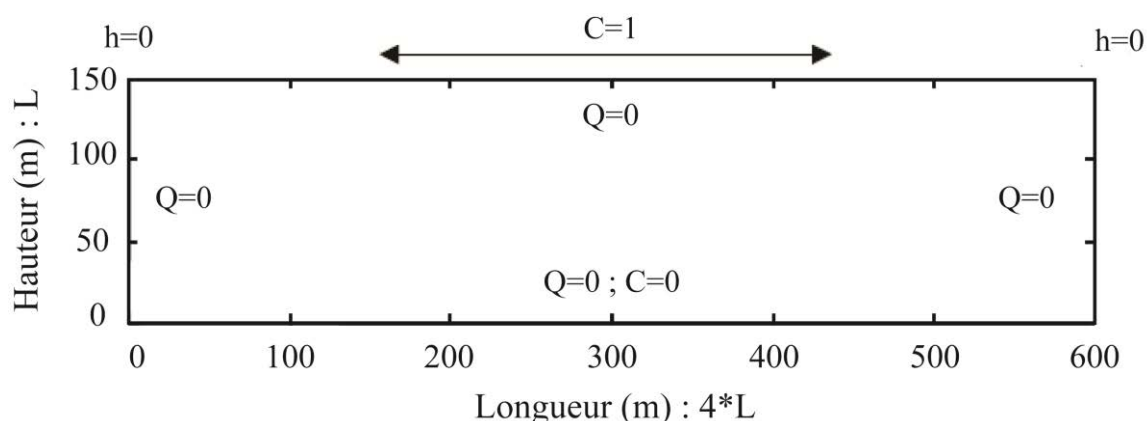


Figure 37. Conditions limites et initiales du problème d'Elder.

Tableau 3. Paramètres de la modélisation pour le problème d'Elder.

Symbole	Paramètre	Valeur	Unité
Dm	Coefficient de diffusion moléculaire	3.565E-06	m ² /s
g	Accélération de la pesanteur	9.81	m/s ²
k	Perméabilité intrinsèque	4.845E-13	m ²
θ	Porosité	0.1	
α	Coefficient de dispersivité longitudinale et transversale	0 ; 0	m
ρ_0	Densité eau douce (Référence)	1000	kg/m ³
ρ_s	Densité soluté	1200	kg/m ³
μ	Viscosité dynamique	1.00E-03	kg/m/s
L	Dimension du modèle	150	m

Avant de présenter les résultats de ces simulations, une courte description théorique du processus mis en jeu peut être faite.

Aspect théorique

Une instabilité hydrodynamique intervient dès lors qu'un fluide plus mobile en déplace un autre. L'interface entre ces deux fluides devient instable et peut donner naissance à des « inter-digitations » ou « fingers » de fluide plus mobile qui progresse au sein du fluide moins mobile. Les propriétés de mobilité des fluides sont reliées à la différence de viscosité et/ou de densité des deux fluides considérés. Des instabilités peuvent naître à l'interface horizontale entre deux fluides de densité différente lorsque le plus dense est situé au-dessus du moins dense (dans la direction du champ de

pesanteur). Ce phénomène est connu sous le nom d'instabilité de Rayleigh-Taylor ou « density fingering ».

Dans un milieu poreux, la vitesse du fluide u est donnée par la loi de Darcy qui varie linéairement avec le gradient de pression ∇p :

$$\nabla p = -u \cdot \frac{\mu}{k} + \rho g$$

La mobilité du fluide est donnée par le coefficient $\frac{k}{\mu}$ ratio entre la perméabilité du milieu poreux et la viscosité du fluide. Considérant le déplacement schématisé de la Figure 38 ou le déplacement se fait dans la direction z , la loi de Darcy s'écrit:

$$\frac{\partial p}{\partial z} = -\frac{\mu}{k} u + \rho g$$

L'interface est instable si une perturbation y est appliquée et que celle-ci grossit. Cette situation se vérifie si la pression à travers la perturbation Δz est supérieure à 0 (Hill et al., 1952; Homsy, 1987) :

$$P_1 - P_2 = -\frac{\mu}{k} \cdot U \cdot \Delta z + (\rho_1 - \rho_2) \cdot \Delta z \cdot g > 0$$

L'instabilité débute donc lorsque, $\rho_1 > \rho_2$, équivalent à la présence d'un fluide plus dense sur un de plus faible densité (cas du problème d'Elder), ou la vitesse moyenne de déplacement est U . Dans ce cas, il existe une vitesse critique de déplacement pour l'instabilité :

$$U_c = \frac{k \cdot (\rho_1 - \rho_2) \cdot g}{\mu} = \frac{K \cdot \Delta \rho}{\rho}$$

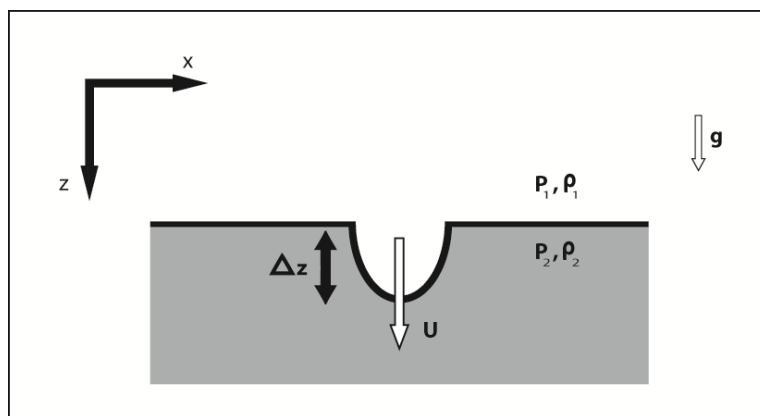


Figure 38. Représentation schématique du déplacement d'une instabilité au sein d'un milieu.

Simulations et résultats

Les simulations réalisées avec COMSOL pour le problème d'Elder ont été réalisées avec les paramètres et les conditions limites décrites précédemment (Figure 37 et Tableau 3). En l'absence de solution exacte pour ce problème, la vérification de la capacité du modèle à reproduire ses effets passe par la comparaison avec les précédentes reproductions de nombreux auteurs (Elder, 1967; Kolditz et al., 1998; Oldenburg and Pruess, 1995; Voss and Souza, 1987). Les résultats obtenus avec COMSOL ont été synthétisés dans la Figure 39 et sont comparés avec les résultats de Kolditz 1998. Néanmoins, la comparaison doit rester qualitative du fait de la forte sensibilité au format de la grille utilisée (Simpson and Clement, 2003). La bonne adéquation entre les lignes d'isoconcentration obtenues à partir des deux simulateurs différents démontre la capacité de COMSOL à reproduire ce type de processus.

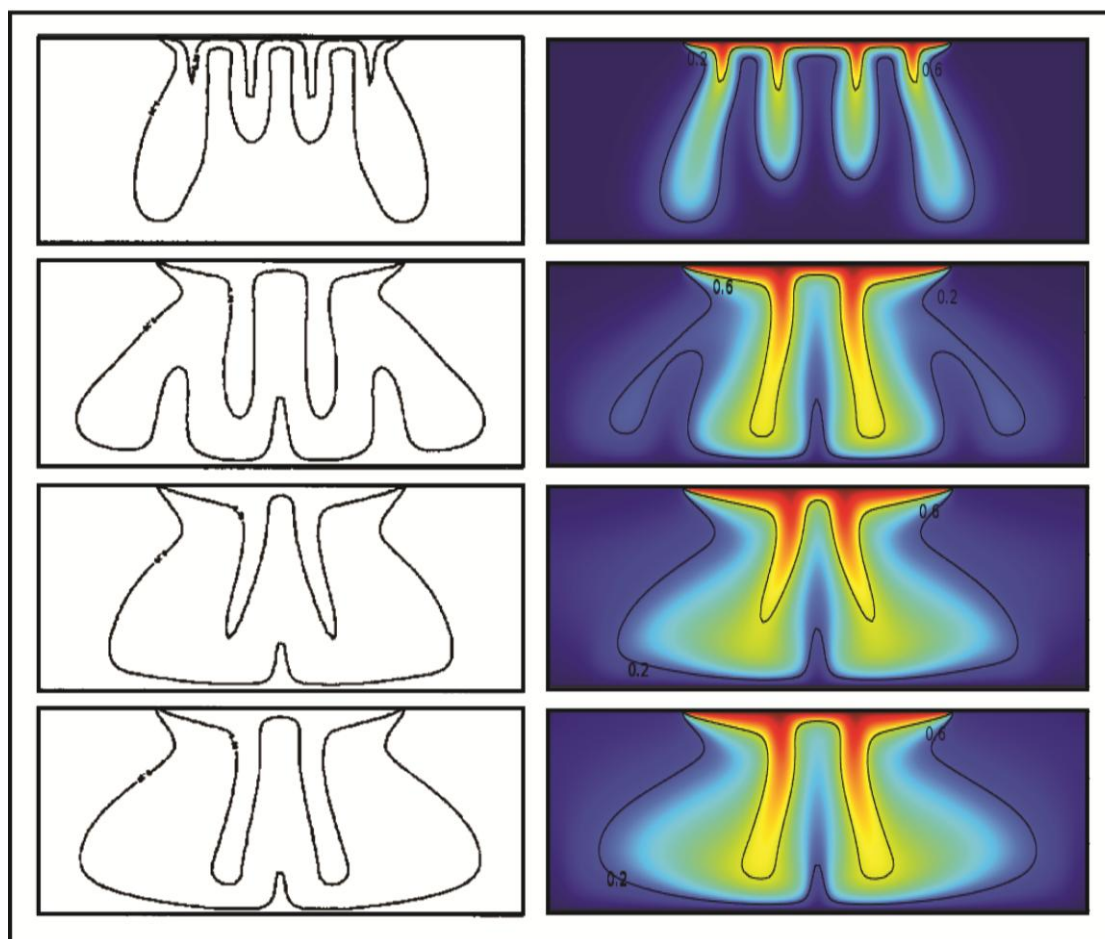


Figure 39. Résultats pour le problème d'Elder: Lignes d'isoconcentration de 0.2 et 0.6 (équivalent à la fraction de concentration du soluté le plus dense) pour des pas de temps de simulation de 4, 10, 15 et 20 ans. À gauche sont présentés les résultats obtenus par Kolditz et al. en 1998 avec une très fine grille. À droite les résultats obtenus avec COMSOL.

« The salt dome problem » (Hydrocoin, level 1, case 5)

Ce problème issu du projet "Hydrocoin project, level 1, case 5" a été développé dans le but de représenter grossièrement le dôme de sel à Gorleben en Allemagne. Ce site reconnu comme pouvant potentiellement accueillir des déchets hautement radioactifs a donné naissance à ce cas-test. Ce problème d'écoulement et de transport en régime permanent est fortement couplé du fait des importantes variations de densité mises en jeu.

Dans ce problème de l'eau douce entre dans le système par la limite supérieure gauche. L'eau douce circule à travers le système en passant par la région salée située en profondeur. Dans cette région le champ d'écoulement est modifié du fait de la différence de densité. Enfin, l'eau ressort par la limite supérieure droite du modèle. Ce problème

correspond donc au lessivage d'un dôme de sel sous l'influence d'une boucle de circulation d'eau douce.

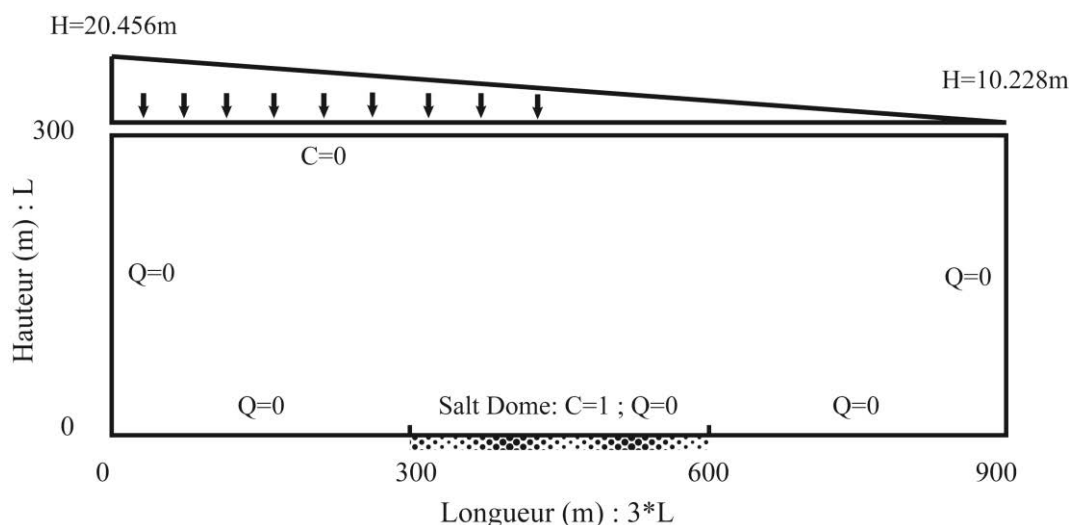


Figure 40. Conditions limites du modèle pour le problème du dôme salé.

Conditions limites et initiales du modèle standard

Le problème du dôme de sel est représenté par un aquifère rectangulaire de 900m de long et de 300m d'épaisseur, considéré homogène et isotrope. La limite inférieure et les limites verticales correspondent à des limites à flux nuls tandis que la limite supérieure est représentée par une variation linéaire de charge hydraulique qui induit l'écoulement au sein de l'aquifère. Le dôme de sel est représenté par une concentration constante localisée au centre de la limite inférieure de 300m de large (pour plus de détails : Herbert, 1988). Au temps $t=0$, le milieu poreux ne contient que de l'eau douce. La géométrie et les conditions limites de ce problème sont synthétisées dans la Figure 40. Les paramètres utilisés dans ce problème sont synthétisés dans le Tableau 4.

Tableau 4. Paramètres de la modélisation pour le problème du dôme salé

Symbole	Paramètre	Valeur	Unité
Dm	Coefficient de diffusion moléculaire	5E-6 à 2.78E-9	m ² /s
g	Accélération de la pesanteur	9.81	m/s ²
k	Perméabilité intrinsèque du milieu poreux	1.0E-12	m/s
θ	Porosité	0.2	
α	Coefficient de dispersivité longitudinale et transversale	0, 0 ou 20, 2	m
ρ	Densité eau douce (Référence)	1000	kg/m ³
S	Densité du soluté	1200	kg/m ³
μ	Viscosité dynamique	8.9E-04	kg/m/s
L	Dimension du modèle	300	m

Simulations et résultats

Dans le passé, cette problématique de lessivage a été analysée par de nombreux auteurs (Herbert, 1988; Konikow et al., 1997, 1996; Oldenburg and Pruess, 1995). Ces auteurs se sont intéressés aux effets de variations des paramètres de diffusion moléculaire et de dispersivité sur le lessivage du dôme salé. C'est pourquoi, ici, différents modèles diffusifs/dispersifs ont été réalisés avec COMSOL afin de comparer les résultats avec les précédentes études (Tableau 5).

Tableau 5. Valeurs des paramètres de diffusion et de dispersivité pour les 4 modèles testés

Modèle	Diffusion moléculaire Dm (m²/s)	Dispersivité longitudinale α (m)	Dispersivité transversale α (m)
1	5.00E-06	0	0
2	5.00E-07	0	0
3	5.00E-08	20	2
4	2.78E-09	20	2

Les résultats obtenus pour les 4 modèles sont synthétisés dans la Figure 41 et comparés avec les résultats des simulations de Herbert (1988). Les différents résultats mettent en évidence l'importance des paramètres du modèle sur la distribution de la salinité et donc plus généralement sur les processus de salinité. La bonne adéquation des résultats entre les modèles diffusifs-dispersifs met en avant la capacité de COMSOL à modéliser les écoulements à densité variable.

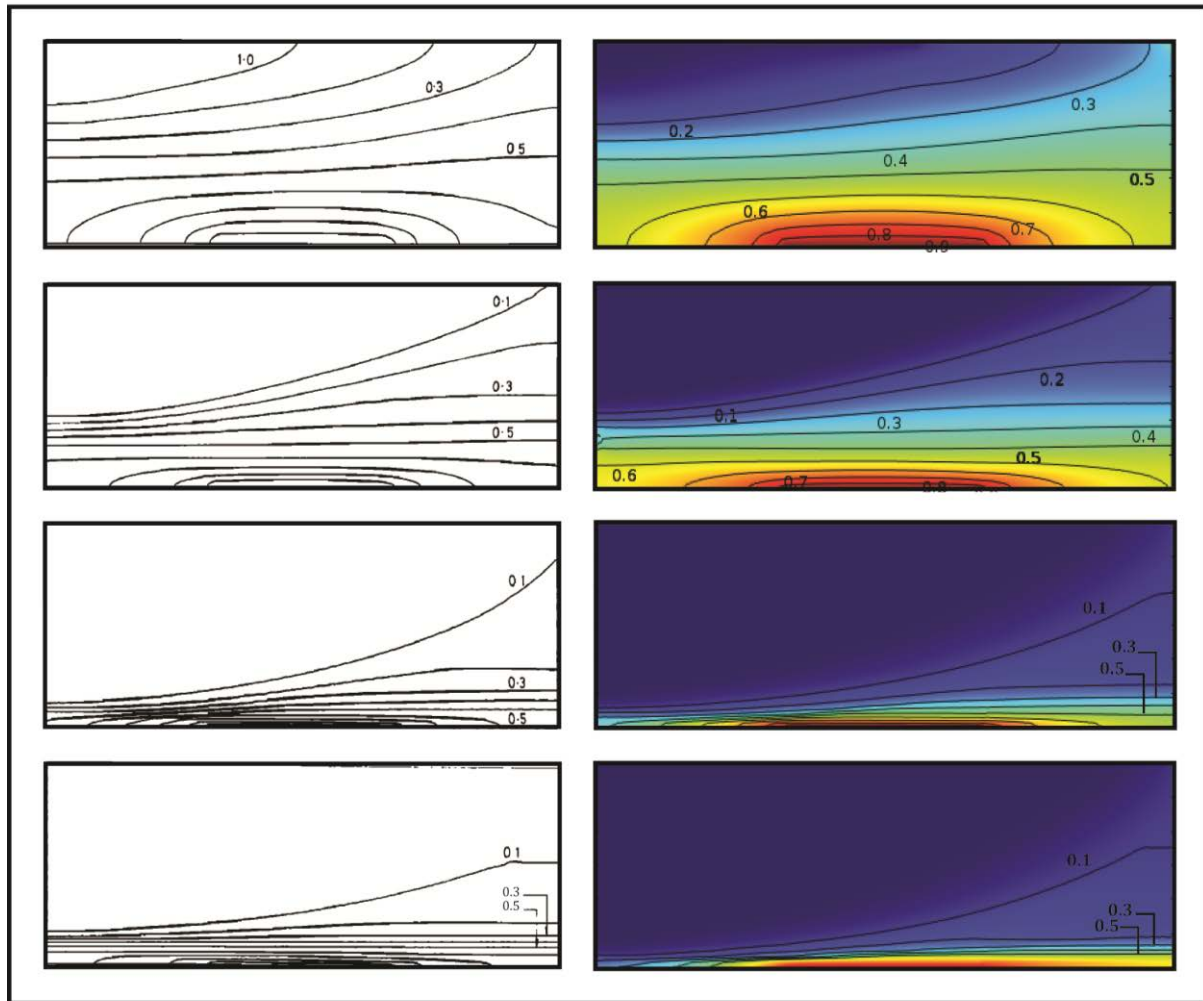


Figure 41. Résultats des simulations en régime permanent pour les 4 modèles. À gauche les résultats des simulations de l'étude d'Herbert 1988. À droite les résultats obtenus avec COMSOL.

Les 3 cas tests réalisés avec le logiciel COMSOL Multiphysics constituent un point de départ dans la réalisation des études qui suivent dans cette thèse. En effet, les questions posées mettent en jeu de l'eau de mer et de l'eau douce ce qui nécessite la prise en compte des écoulements à densité variable.

4.3. *Downconing of saline water below coastal rivers*

Article submitted to (/2014)

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Keywords: Gravity instability; salt intrusion ; river ; groundwater

ABSTRACT

This study investigated the salinization of continental aquifers by the process of gravity instability below coastal rivers. Due to connection with the sea a leading edge of seawater may enter deeply inland through fluvial systems. The occurrence of the salt front enables the presence of seawater (or fraction of seawater) on a fresh groundwater system and can cause seawater intrusion by gravity instability. This is a general phenomenon taking place in all fluvial systems connected to the sea, and hence can cause widespread salinization. Our study used numerical experiments to investigate salinization of fresh groundwater by salinized rivers. The system investigated involves a 2-dimensional vertical cross-section drawn perpendicular to the river with an underlying saturated porous medium. Modeling considers three main parameters: (1) the groundwater system permeability, (2) the salinity of water in contact with the river bed and (3) the river width. Individual relationships established between these parameters and the salt water inflow rate provide a basis for a general expression. The general expression and the critical hydraulic gradient will enable to produce regional risk map and an assessment of groundwater salinization rates at large scale according to geomorphological and environmental characteristics of the region. The findings could be applied in the context of climate change, which is expected to lead to sea-level rise. Management of groundwater resources in coastal areas will have to address this major issue in the near future.

1. INTRODUCTION

Seawater intrusion in groundwater systems is characterized by the landward movement of seawater into fresh groundwater (salinization). Seawater intrusion most frequently occurs in coastal regions due to their proximity to seawater and the intensive water needs that occur with population pressures. Salinization mainly results from prolonged variations of hydrological conditions in coastal and estuarine regions in response to natural processes, including climate variations, geological coastal evolution or sea-level fluctuations (Carretero et al., 2013; Chang et al., 2011; Gossel et al., 2010; Maas, 2007; Sherif and Singh, 1999; Watson et al., 2010; Webb and Howard, 2011; Werner and Simmons, 2009), episodic events such as tsunamis (Illangasekare et al., 2006; Violette et al., 2009) and storms (Terry and Falkland, 2009; Wilson et al., 2011), interactions with surface water bodies (recharge variations...)(County et al., 2004; Oude Essink et al., 2010) and sea dynamics, such as tidal effects (Ataie-Ashtiani et al., 1999; Werner and Lockington, 2006). However, anthropogenic activities such as groundwater abstraction, urbanization, land reclamation and drainage, may disturb the hydrological systems and may also influence the salinity distribution (Calvache and Pulido-Bosch, 1997; Jakovovic et al., 2011; Luyun et al., 2011; Pool and Carrera, 2011; Reilly and Goodman, 1987). These main processes (natural and anthropogenic) and driver factors in coastal environments associated with seawater intrusion have been described qualitatively by Custodio et al., (1987) and reported in exhaustive reviews (Bear et al., 1999; Werner et al., 2013). Nevertheless, the interactions between processes provide an infinite array of seawater intrusion settings.

Seawater intrusion can occur through different pathways depending on the density distribution and more specifically to the relative distribution of freshwater and seawater within the groundwater system, considering that the primary driver factor of seawater intrusion are buoyancy forces associated with density variations. The main pathways include lateral intrusion of seawater, downwards seepage from a saline surface water body that overlies an aquifer and upward intrusion from deeper more saline strata as up-coning of deep saline seawater (Barlow, 2003; Pattle Delamore Partners Ltd, 2011).

The most common groundwater salinity distribution in aquifers corresponds to a stable density stratification where fresh water overlies saline water, thus creating an interface called the mixing zone. The fluid density variation (between seawater and freshwater) through the mixing interface is an important characteristic of coastal aquifers. This process contributes to the convective circulation within the wedge and hence drives seawater intrusion into the freshwater discharge (Abarca et al., 2007a, 2007b) and can extend over a few meters to several kilometers. In first approximation, the Ghyben-Herzberg (Ghyben, 1889; Herzberg, 1901) principle is used to provide an estimate of the saltwater interface depth but it is more frequently represented by the Henry problem (Henry, 1964). The lateral intrusion, the mixing zone thickness and its various configurations are represented by the Henry problem and have been studied through laboratory experiments, field situations but predominantly investigated with numerical modeling (Abarca, 2006; Henry, 1964; Pinder and Cooper, 1970; Segol, 1994; Simpson and Clement, 2004; Voss and Souza, 1987; Werner et al., 2013). This process is driven by mechanical dispersion, molecular diffusion, freshwater discharge and the density contrast (Abarca and Clement, 2009; Barlow, 2003; Goswami and Clement, 2007; Held et al., 2005; Jakovovic et al., 2011; Paster and Dagan, 2007; Volker and Rushton, 1982). However, heterogeneities may have an impact on the mixing zone phenomenon as noticed by several authors (Abarca et al., 2007a; Held et al., 2005; Kerrou and Renard, 2010; Rahman et al., 2005).

Geological properties of the groundwater system (anisotropy, degree of confinement, layers...) involve variations in hydraulic properties and have an impact on the transport of dissolved constituents (Diersch and Kolditz, 2002). The distribution of geological properties can provide preferential seawater intrusion pathways (Calvache and Pulido-Bosch, 1997; Mulligan et al., 2007). These preferential flow paths may induce unstable density configurations because of the setting of seawater on fresh groundwater which can cause downward vertical seawater intrusion (Diersch and Kolditz, 2002; Simmons et al., 2001; Xie et al., 2011). This configuration characterized by downward movements and in some cases by free convection through instabilities, can be the result of heterogeneity within the groundwater system or external events that bring seawater over fresh groundwater: transgressions (e.g. Kooi et al., 2000), tsunamis (e.g. Illangasekare et al., 2006) and storms (e.g. Anderson Jr. and Lauer, 2008). Unstable convective flows have been the focus of many studies for almost a century and continue

to be of interest for the international community (Elder, 1967; Rayleigh, 1916; Xie et al., 2012).

Seawater intrusion processes in coastal regions have received considerable interest from the international scientific community over the last decades (Abarca, 2006; Barlow, 2003; Bear et al., 1999a; Custodio et al., 1987; Henry, 1964; Pinder and Cooper, 1970; Werner et al., 2013). Nevertheless, there is a lack of specific studies on possible processes that can take part in significant groundwater salinization. As mentioned by Werner et al. (2013), indirect effects such as salinity increase in surface water bodies due to seawater penetration can contribute significantly to seawater intrusion. Thus, we investigate how that groundwater salinization may originate from downward seepage from salinized coastal surface bodies, as rivers. Due to high density of rivers connected to the sea and seawater incursion deeply inland through these fluvial systems, rivers may be a major player of salinization of continental aquifers.

Nowadays, the presence of the salt front (the leading edge of seawater entering the fluvial system due to connection with the sea) further inland (more than several tens of kilometers) is well known. The river salinity evolves with the distance from the sea, with seawater salinity (35g/L, see Millero et al., 2008) in estuary and a small fraction at several tens of kilometers inland. Figure 42 shows the steady-state salinity profile of the Loire, one of the main rivers in France which flows into the Atlantic Ocean. Salinity is visible approximately up to 50 km from the sea (SYVEL database: <http://www.loire-estuaire.org/dif/do/init>).

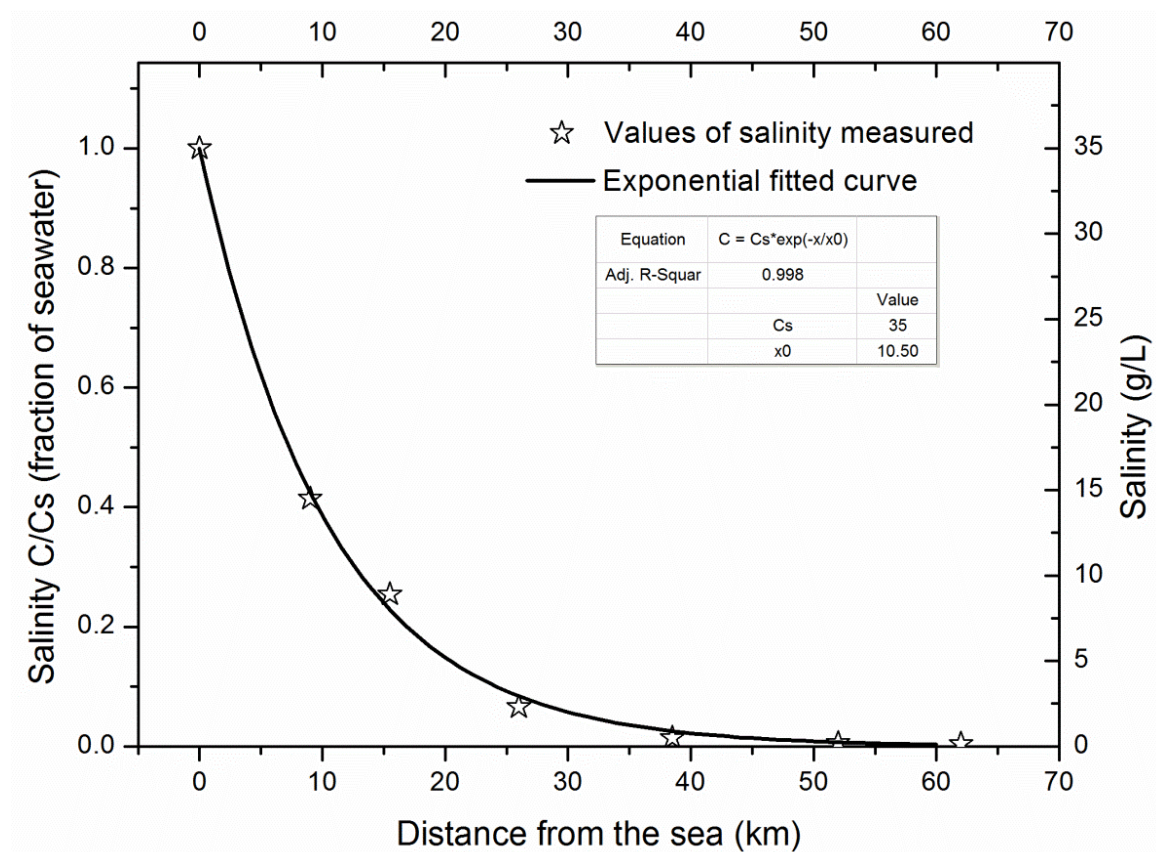


Figure 42. Salinity profile of the Loire as function of the distance from the sea

Thus, the occurrence of seawater (or fraction of seawater) on fresh groundwater exists due to the presence of the salt front inland and can cause seawater intrusion by gravity instabilities as schematized in Figure 43. Moreover, this is a global phenomenon which takes place at large scale in all fluvial systems connected to the sea (Ho et al., 2002; Monismith et al., 2002; Wollast and Peters, 1978), and hence can cause widespread salinization.

The aim of this work is to focus on the groundwater salinization processes occurring in coastal fluvial system depending on the width, the salinity of the stream and properties of the groundwater system.

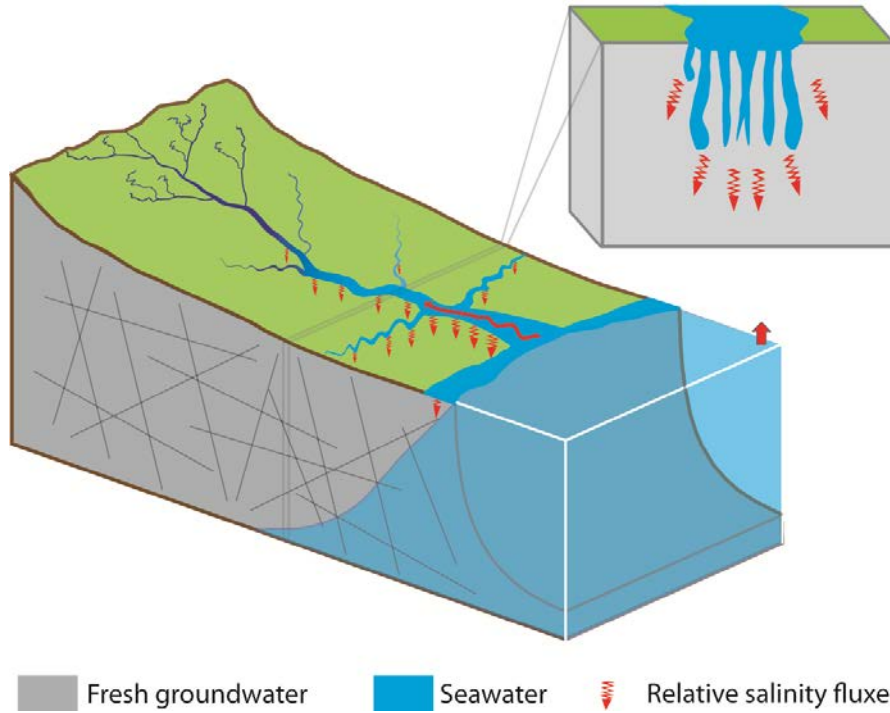


Figure 43. Conceptualization of the salinization process occurring below fluvial systems

2. NUMERICAL METHODS AND PARAMETERS

2.1. INSTABILITIES PHENOMENA

Figure 43 provides an idealized cross-section of a river belonging to a fluvial system influenced by seawater overlying a fresh groundwater system. This density stratification where seawater (or fraction of seawater) is on top of a fresh groundwater domain is the main reason of instability phenomena causing seawater intrusion. In fact, when the density contrast is sufficiently high, lobes of dense fluid form and move downward (fingering), counterbalanced by upward movements of less dense fluid (Wooding et al., 1997). The onset of instabilities can be characterized by the Rayleigh number that provides the instability degree in the porous media and the onset of free convection. The dimensionless Rayleigh number is defined as:

$$Ra = \frac{\Delta \rho \cdot K \cdot H}{\rho_0 \cdot \theta \cdot D_m} = \frac{V_s \cdot H}{D_m}$$

where $\Delta \rho$ is the density contrast between the seawater and the freshwater density [$\text{kg} \cdot \text{m}^{-3}$], K is the hydraulic conductivity of the porous domain, ρ_0 is the density of freshwater, θ is a dimensionless porosity, D_m is the molecular diffusion coefficient

$[m^2.s^{-1}]$, H is a characteristic height of the porous domain and V_s is the theoretical velocity of instability.

When the Rayleigh number is higher than the critical Rayleigh number Ra_c , convective transport with density-driven fingers (or plumes) takes place (Simmons et al., 2001). First, Ra_c has been estimated at 4×10^9 (Lapwood, 1948) for specific conditions (infinite, horizontal, uniform porous layer with no-flow and constant temperature boundaries), but this critical value has been recently reviewed by van Reeuwijk et al., 2009 where $Ra_c = 0$ for the onset criterion of free convection. Situations where $Ra \gg 0$ underline that for a small fraction of seawater in river free convection can occur, and highly unstable conditions can be expected in the porous media. It can be noted that molecular diffusion plays a negligible role in the process.

The theoretical free convective fingering velocity (V_s) is approximated by a theoretical speed building on hydrogeologic parameters of the considered system (permeability, porosity...) from the flux u according to Darcy's law, obtained as follows:

$$u = -\frac{k}{\mu} (\nabla p - \rho g)$$

where u is the volumetric fluid flux $[m.s^{-1}]$, ρ is the density of the fluid $[kg.m^{-3}]$, k is the intrinsic permeability $[m^2]$, μ is the dynamic viscosity of the fluid $[kg.m^{-1}.s^{-1}]$, p the fluid's pressure $[Pa]$, g the acceleration due to gravity $[m.s^{-2}]$ and ∇ is the divergence operator.

Gravity instability occurs due to the density contrast between seawater and freshwater within a homogenous and isotropic porous domain and the flux u obtained from Darcy's law is related to pore velocity V are related to by $u = \theta.V$, the theoretical velocity of instability can be written as:

$$V_s = \frac{k \cdot \Delta \rho \cdot g}{\theta \cdot \mu} = \frac{K \cdot \Delta \rho}{\theta \cdot \rho_0}$$

Understanding the instability velocity has been the focus of numerous studies (Gebhart et al., 1988; Kooi et al., 2000; Riaz et al., 2006; Stevens et al., 2009; Wooding et al., 1997) and have been recently summarized and further studied (Xie et al., 2011). Nevertheless, the real instability velocity is slower than (V_s) due to dispersion-diffusion and upwelling

effects. The real fingering speed V_g [m.s⁻¹] in a system needs to be calculated with a correction factor f_c [dimensionless] with a value between 0.115 and 1. The real speed of gravity instabilities can be expressed as:

$$V_g = f_c \cdot V_s$$

Further investigations are required for adjusting this theoretical velocity in order to include significant parameters, such as dispersion (Xie et al., 2011).

Quantification of saline fluxes beneath a salinized river requires to couple equations for water and salt transport. These equations are strongly coupled due to the importance of fluid density. To study the behavior of this salinization process, a linearized form of Darcy's law for fluid flow in porous media is used with the conservation equations for fluid and salt.

2.2. NUMERICAL SIMULATOR AND GOVERNING EQUATIONS

Numerical experiments reported in this study were carried out with COMSOL Multiphysics. COMSOL is a finite element numerical simulation program used to solve physics and engineering problems, specifically coupled phenomena. The software performance to resolve variable-density flow and transport was demonstrated by solving the classical benchmark problems (Elder, 1967; Li et al., 2009). For a detailed description of the software and the code, readers can refer to the COMSOL Multiphysics Modeling Guides (<http://www.comsol.com/shared/downloads/4.3b/ReleaseNotes.pdf> and <http://nf.nci.org.au/facilities/software/COMSOL/4.3/doc/pdf/mph/COMSOLMultiPhysicUsersGuide.pdf>). COMSOL allows the solution of two coupled non-linear partial differential equations through a 2-way coupling of two physics interfaces: Darcy's law representing flow in porous media and advective-dispersive solute transport equation. The software COMSOL solves the governing equations of the mass-balance for fluid and solute. These governing equations are briefly explained below.

The equation of conservation of the mass of fluid can be written with Darcy's law:

$$\frac{\partial(\rho\theta)}{\partial t} + \nabla \cdot (\rho \mathbf{u}) = 0$$

For the solute, represented by a single conservative dissolved species stored in a solution, the mass conservation equation can be given as:

$$\frac{\partial(\theta\rho C)}{\partial t} = \nabla \cdot (\theta\rho D \cdot \nabla C) - \nabla \cdot (\theta\rho u C)$$

where u is the volumetric fluid flux obtained from Darcy's velocity [m.s^{-1}], ρ is the density of the fluid [kg.m^{-3}], θ is a dimensionless porosity, ∇ is the divergence operator, t is the time [s], k is the intrinsic permeability [m^2], μ is the dynamic viscosity of the fluid [$\text{kg.m}^{-1}.\text{s}^{-1}$], p the fluid's pressure [Pa], C is the solute concentration [kg.m^{-3}] and D is the hydrodynamic dispersion coefficient [$\text{m}^2.\text{s}^{-1}$].

The coefficient D is defined as the effect of two combined processes, the mechanical dispersion and molecular diffusion (J Bear, 1972; Scheidegger, 1961) and can be expressed as:

$$D = D_m + \alpha_v |u| + (\alpha_L - \alpha_v) \frac{u^2}{|u|}$$

where D_m is the molecular diffusion coefficient [$\text{m}^2.\text{s}^{-1}$] and α_L , α_v the intrinsic longitudinal and transversal dispersivity, respectively [m].

The equations are coupled due to the density which is used to calculate the two mass-balance equations in relation to both fluid flow and mass transfer: fluid density appears in both equations and it is dependent on solute concentration. Then, the concentration is used to calculate the density of the fluid and a linear equation is used to relate solute concentration to density.

Over a limited range of salinity, it can be assumed that density is linearly proportional to concentration (Frind, 1982; Henry, 1964; Voss and Souza, 1987):

$$\rho = \rho_0 \cdot \left(1 + \beta \cdot \frac{C - C_0}{C_s - C_0}\right)$$

where ρ_0 is the density of the reference fluid (freshwater) [kg.m^{-3}] corresponding to the initial concentration C_0 , C is the solute concentration which contributes to the density variation [kg.m^{-3}], C_s is the solute concentration with the maximum density (seawater) [kg.m^{-3}] and β is the density difference ratio defined as:

$$\beta = \frac{\rho_s - \rho_0}{\rho_0} = \frac{\Delta\rho}{\rho_0}$$

2.3. MODEL GEOMETRY AND BOUNDARY CONDITIONS

Numerical model experiments have been carried out with a 2-dimensional vertical cross-section drawn perpendicular to the river. This geometry was chosen to be consistent with the conceptual model (Figure 43). The geometry and boundary conditions applied in this study are described in Figure 44. Here, we considered a groundwater system with a width of 2 km and a depth of 1 km. The large scale was chosen to be more representative of natural settings and to observe the salinization process on the long term. At the top boundary, a constant concentration boundary condition with a unit relative to seawater concentration ($\frac{C_{river}}{C_s}$) is localized at the middle of the top side according to a width (W) to act as a salinized river. The maximum concentration ($C = 1$) of overlying dense water is 1025 kg.m^{-3} , which is equivalent to seawater salinity within river. The entire top boundary is modeled with a fixed head (0m) boundary condition and on each side of the river a zero concentration value ($C = 0$) is applied. The left, bottom and right sides were assigned to no flow boundary condition. At initial conditions, the entire model domain was saturated with pure fresh groundwater ($C = 0$).

As a first approximation, simulations do not include the active groundwater flow which feeds the river (fixed head (0m) at the entire top boundary). In fact, in the most natural cases, streams receive water from groundwater discharges which occur in horizontal and vertical directions to the stream and hence are in opposition with the direction of free convection process. Thus, as a first step, it seems reasonable to consider that this study provides a reasonable estimation of groundwater salinization below rivers influenced by seawater in low-lying areas with limited topography on each side of a coastal river.

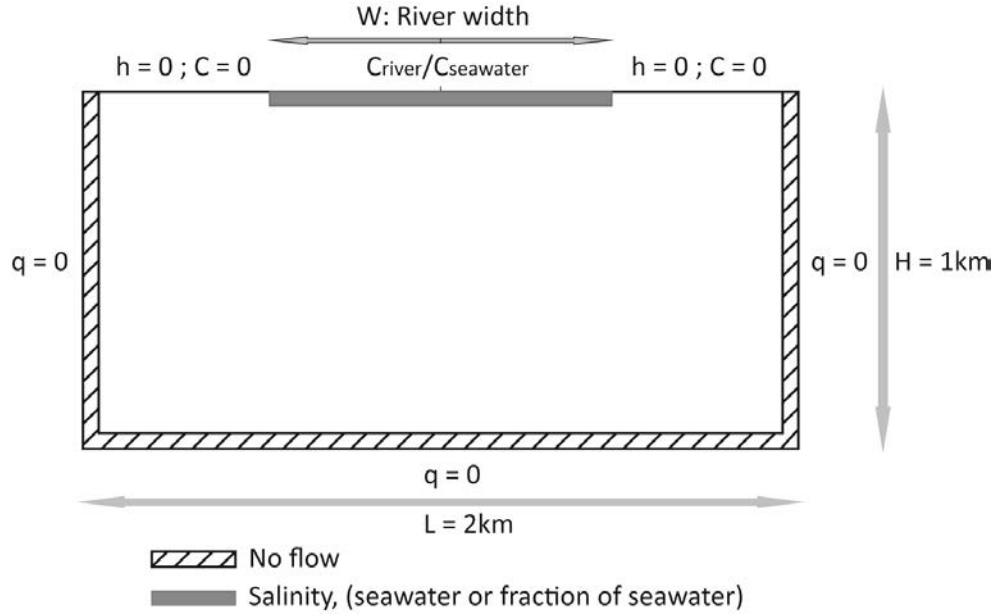


Figure 44. Model geometry, boundary and initial conditions for simulations.

Then, as a second step, to take into account the groundwater flow which feeds fluvial system would be necessary because it may affect salinization efficiency. Seawater inflow rate may be limited proportionally to the groundwater fluxes.

Fresh groundwater velocity is generally expressed as:

$$V_f = \frac{K}{\theta} \cdot i$$

where V_f is the average velocity of fresh groundwater [m.s^{-1}], K the permeability of the groundwater system [m.s^{-1}], θ the porosity [dimensionless] and i the hydraulic gradient [dimensionless]. And, as stated previously, the velocity of gravity instability corresponds to:

$$V_g = f_c \cdot \frac{K}{\theta} \cdot \frac{\Delta}{0}$$

Assuming that salinization process is inefficient when:

$$V_g \leq V_f$$

$$f_c \cdot \frac{\Delta}{0} \leq i$$

This above condition suggests that groundwater systems which are influenced by gravity instability below salinized rivers are dependent on the salinity degree of the river and the hydraulic gradient and therefore directly correlated to the topography close to the river. Indeed, hydraulic gradient corresponds to a height variation (Δh_{Topo}) over a distance (L).

$$i = \frac{\Delta h_{\text{Topo}}}{L}$$

2.4. PARAMETRIC NUMERICAL EXPERIMENTS

Considering the parameters used for the numerical experiments of this study listed in Table 1, the actual natural convection system is characterized by $Ra \gg 0$ which implies that free convection processes dominate salt transport. The investigation of salinization process in continental aquifers below rivers according to the complexity of natural systems environments is studied in response to three major parameters: the river width (W), the salinity of the river (C_{river}) and the groundwater system permeability (K).

Natural fluvial systems are typically characterized by wide ranges in river width (W). To observe the effect of channel width on salinity inflow rate into fresh groundwater system, simulations have been carried out with a river concentration ($\frac{C_{\text{river}}}{C_s} = 1$) fixed and a range of river width ($W = 1 \text{ to } 1000 \text{ m}$) which varies on four orders of magnitude (see Table 1). Here we have investigated the importance of the channel width relative to the aquifer system size.

As stated previously, river salinity evolves with the distance from the sea due to the leading edge of seawater entering the fluvial system (Figure 42). In the estuary seawater occurs with a density around 1025 kg.m^{-3} ($C=1$) and the proportion of seawater evolves until to the fresh water salinity and density of 1000 kg.m^{-3} is reached (at several kilometers from the sea). To quantify the effect of river concentration on salt water inflow rate, simulations were done with a river width ($W = 100 \text{ m}$) fixed and a range of concentration at the river boundary. Where, concentration is expressed as fraction of seawater concentration (1, 0.5, 0.25 and 0.1) (Table 1).

Table 1. Model parameters used in simulations. Unchanged parameters are parameters held constant for all simulations. Tested parameters correspond to values of the three major factors tested in this study

Unchanged parameters				
Fresh groundwater density (ρ_0)	1000	kg.m ⁻³	Longitudinal permeability (α_L)	1 m
Initial fresh groundwater concentration (C_0)	0	kg.m ⁻³	Transverse dispersivity (α_v)	1 m
Seawater density (ρ_s)	1025	kg.m ⁻³	Molecular diffusivity (D_m)	2.10 ⁻⁹ m ² .s ⁻¹
Maximum density difference ($\Delta\rho$)	25	kg.m ⁻³	Acceleration due to gravity (g)	9.81 m.s ⁻²
Fluid dynamic viscosity (μ)	10 ⁻³	kg.m ⁻¹ .s ⁻¹	Aquifer depth (H)	1000 m
Porosity (θ)	0.1		Aquifer Length (L)	2000 m
Tested parameters				
Groundwater system permeability (K) [m.s ⁻¹]	$K_0, K_1, K_f, K_{f1}, K_{f2}$		with $W=100m$ and $C_{river}/C_s=1$	
River width (W) [m]	10, 50, 100 1000		with K_f and $C_{river}/C_s=1$	
River concentration (C_{river}) [C_{river}/C_s]	0.1, 0.25, 0.5, 1		with K_f and $W=100m$	

where $K_0 = 5.10^{-6}$; $K_1 = 5.10^{-7}$; $K_f = 5.10^{-6} .e^{-z/0.25km}$; $K_{f1} = 5.10^{-6} .e^{-z/0.125km}$; $K_{f2} = 5.10^{-6} .e^{-z/0.025km}$

Natural groundwater systems are generally characterized by various organizations and wide range of permeability (K), resulting in a wide spectrum of Ra and thereby in numerous degrees of instability. To constrain the impact of permeability of the groundwater system on salinity fluxes, simulations have been computed with a river concentration ($\frac{C_{river}}{C_s} = 1$), a river width ($W = 100 m$) and several permeability vertical distributions (Table 1). First, two homogeneous models have been tested with constant permeability $K_0 = 5.10^{-6} m.s^{-1}$ and $K_1 = \frac{K_0}{10} = 5.10^{-7} m.s^{-1}$, (Figure 45) in accordance with the initial permeability of the exponential relationship used for the continental crust. Typical permeability model (exponentially decreasing with depth) have been proposed to represent the continental crust (Saar and Manga, 2004). They propose an exponential relationship, $K_f = K_0 . e^{-\frac{z}{z_0}} = 5.10^{-6} . e^{-\frac{z}{0.25km}}$, for continental crust in general and for “shallower” depths (typically $z \leq 0.8km$ and up to $z \leq 2km$). However, for depths $z \geq 0.8km$, their results are in agreement with the power law relationship $K = 10^{-7} . \left(\frac{z}{1km}\right)^{-3.2}$ suggested by Ingebritsen and Manning in 1999. Here, we used the initial exponential relationship (K_f) and two other models generated from this relationship (K_{f1} and K_{f2}) where the characteristic depth (z) is modified $z_1 = \frac{z_0}{2} = 0.125 km$ and $z_2 = \frac{z_0}{10} = 0.025 km$, respectively (Figure 45).

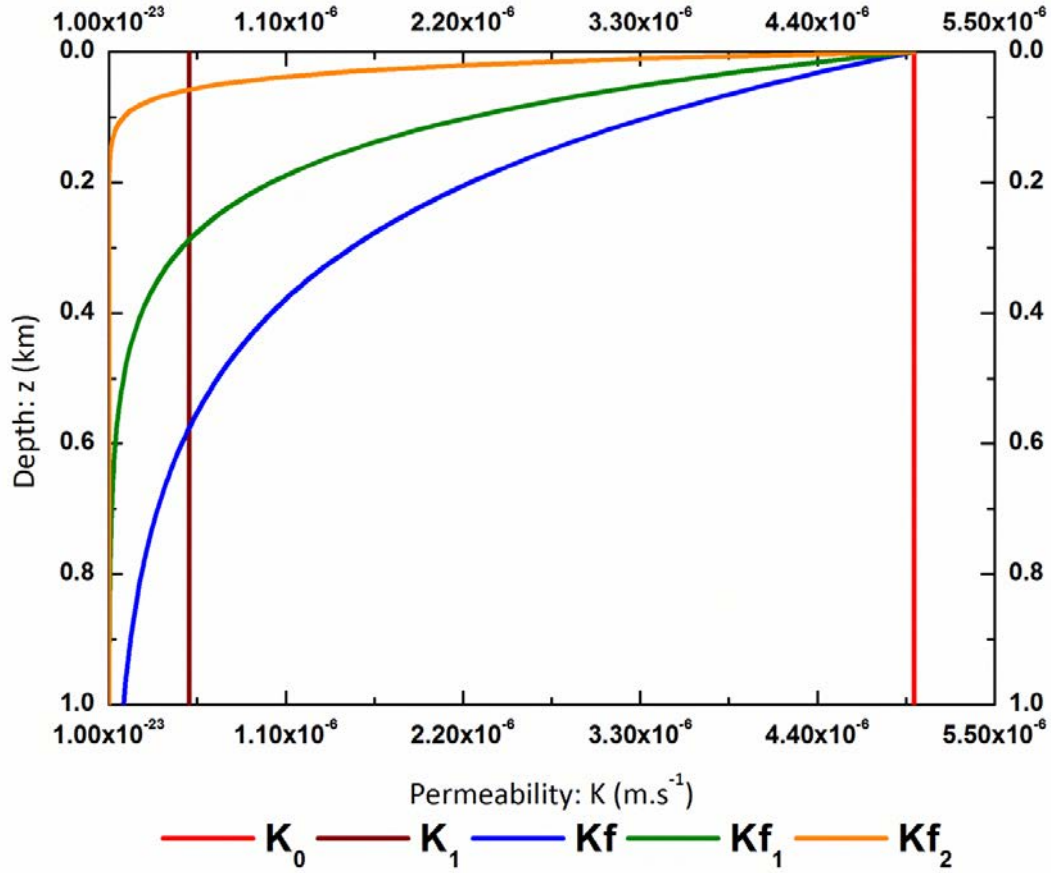


Figure 45. Permeability distributions used in simulations

2.5. SPATIAL AND TEMPORAL DISCRETIZATIONS

To minimize numerical errors during simulations, conditions on mesh size and time step need to be respected.

The mesh Péclet number was defined to determine grid discretization (Diersch and Kolditz, 2002):

$$Pe_m = \frac{|u| \cdot h}{D_m + |u| \cdot \alpha_L} \approx \frac{h}{\alpha_L} \quad 2$$

where α_L is the longitudinal dispersivity and h is the maximum distance between two sides of an element in the direction of the groundwater flow. The maximum element size applied in our simulations is 2 m where gravity instabilities take place. The domain was been re-meshed between steps of simulation according to the location of instabilities to save time and computing capacity. The importance of this parameter was widely reported and so largely used in numerous studies (Graf and Therrien, 2005; Soto Meca et al., 2007; Xie et al., 2011, etc...).

The Courant-Friedrichs-Lewy (CFL) number is defined as:

$$CFL = \frac{|u| \cdot \Delta t}{h} \leq 1$$

To achieve the CFL convergence criteria an initial time step of approximately 100 s was used. The maximum time step was adopted throughout numerical experiments to respect the CFL condition. In COMSOL Multiphysics an implicit method (BDF Backward Differentiation Formula) was applied for the time stepping increment. The maximum time step time was restricted by the minimum element size (h) along the river boundary and the maximum convection speed.

2.6. INITIAL PERTURBATIONS

It is well known that gravity instabilities are initiated by heterogeneities (geologic and/or hydraulic) of the groundwater system (Moissis and Wheeler, 1990) but are still extremely difficult to predict and quantify. In modeling practice the initiation of instabilities is usually the result of numerical instabilities coming from specificities of the source code (Schincariol et al., 1994). Numerical studies have demonstrated by comparison with laboratory results the need to consider initial perturbations at the boundary to have better match (Post and Simmons, 2010; Simmons et al., 1999). In our simulations an initial perturbation was applied to the top concentration boundary representing the river for early times to simulate free convective behavior. The function is described as a parametric curve in COMSOL Multiphysics:

$$A \cdot \sin \left(x(W) \cdot \frac{2\pi}{\lambda} + \frac{\pi}{2} \right)$$

where A is the amplitude [m], $x(W)$ is a distance function of the river width (W) [m] and λ is the wavelength of the initial perturbation [m]. The domain designed from this function is assigned with the concentration applied at the river boundary. Two criteria have been changed for initial perturbation (amplitude and wavelength). For each river width and river concentration early stages of simulations have been tested with several wavelengths λ ranging from 3.125m to 100m. A comparison of results was carried out in order to keep the most efficient configuration for long term simulations. Studies on this topic (List, 1965; Schincariol et al., 1994; Simmons et al., 2001) have shown that most wavelengths are unstable when density difference increases and that the critical

wavelength to perturb the system is smaller. However, gravity instabilities occurring in a groundwater system can be characterized by many solutions (Xie et al., 2011) due to the density effect, characteristic parameters (fluid and groundwater system properties) and boundary conditions of the model. Initial perturbations have an impact on salinity fluxes at the beginning of the salinization process and its influence tends to decrease with time until it reaches a characteristic inflow rate. In this study we focus on salinity fluxes into a groundwater system on long term as function of width, salinity of the river and of groundwater system properties.

3. RESULTS

The numerical experiments have been designed (section 2) to investigate groundwater salinization efficiency below coastal rivers. The quantitative analysis of salinity fluxes into groundwater was carried out relative to three major factors: aquifer permeability (K), river width (W) and river water concentration (C_{river}). In this study, we used a measurable quantity for characterizing salinity inflow rate (q) which corresponds to the average concentration of the groundwater domain over time normalized by seawater concentration and the domain area:

$$q = \frac{A}{C_s} \cdot \frac{d\bar{C}}{dt}$$

where q is the normalized mass flux [$m^2.s^{-1}$], C_s is the seawater concentration [$kg.m^{-3}$], $A = L.H$ is the domain area [m^2] and \bar{C} the average concentration [$kg.m^{-3}$]. At the beginning the average concentration of the domain is null ($\bar{C} = C_0$) and evolves as a function of the salinization efficiency.

3.1. NORMALIZED MASS FLUX AS FUNCTION OF THE GROUNDWATER SYSTEM PERMEABILITY

To investigate the impact of the groundwater system permeability on salinization efficiency, simulations have been carried out with several vertical distributions of permeability (Figure 45) a river concentration ($\frac{C_{river}}{C_s} = 1$, equivalent to seawater) and a river width ($W=100m$) fixed (Table 1).

3.1.1. HOMOGENOUS PERMEABILITY

First, two models have been simulated with constant permeability ($K_0 = 5.10^{-6} m.s^{-1}$ and $K_1 = 5.10^{-7} m.s^{-1}$) where K_0 constitutes the reference model. Figure 46 illustrates the plume patterns from all permeability cases (K_0, K_1, K_f, K_{f_1} and K_{f_2}) for three time steps (5, 50 and 100 years). The reference case (K_0) shows that this system is clearly unstable with fingers of saline water sinking into the fresh groundwater domain under the influence of gravity. At early time, small fingers initiate from the river boundary condition. Then, fingers progress downward and tend to coalesce to constitute two larger fingers which reach the bottom of the groundwater domain. Later, saline fluid fingers spread over the bottom of the aquifer (Figure 46: K_0). In the case (K_1), the system is still unstable with fingers, nevertheless the fingers downward motion is more limited due to the lower convective velocity (Figure 46: K_1) which depends on the permeability. These homogenous cases allow comparison with previous studies and a verification of theoretical values such as characteristic convective velocity studied by several authors (Post and Kooi, 2003; Xie et al., 2011). Convective velocity is computed with the theoretical speed presented previously: $V_s = 1.2.10^{-6} m.s^{-1}$ according to parameters defined in Table 1 and considering the permeability K_0 . This calculated velocity implies that gravity instabilities would reach the bottom of the domain after 26 years which is not the case (see Figure 46: K_0). Results show that instabilities reach the bottom after 50 years and specifically at 62.5 years (Figure 46: K_0). The speed of gravity instabilities is smaller than theoretical value because of dispersion-diffusion and upwelling effects, as demonstrated in previous studies. Theoretical speed needs to be corrected by a corrective factor f_c with a value ranging from 0.115 - 1.000, a constrained range proposed by Xie et al., (2011). For the K_0 simulation, $f_c = 0.4$ which is consistent with the constrained range. To check the corrective factor value $f_c = 0.4$, we used this value to estimate the real instability velocity for K_1 , which gives $V_g = 5.10^{-8} m.s^{-1}$ and suggests that instabilities would have been traveled on approximately 160 m after 100 years. Result on Figure 46 attests this assumption and validates $f_c = 0.4$.

Figure 47 illustrates normalized mass flux (q) for all permeability models. For the reference case (K_0), inflow rate evolves over time with a rapid increase followed by stabilization. The first phase appears to correspond to the setting of instabilities phenomenon (at early time with small fingers, Figure 46: K_0). Then, the system

development continues until reaching a more efficient configuration (larger fingers at later time, Figure 46: K_0) where normalized mass flux reaches the maximum value (Figure 47). In K_1 case, the evolution is quite similar but normalized mass flux value is lower and the time to reach maximum normalized mass flux requires more time (Figure 47). Thus, salinity normalized mass flux is clearly dependent on groundwater system permeability.

3.1.2. EXPONENTIAL PERMEABILITY DISTRIBUTION

In addition to homogeneous cases, permeability models have been computed from a typical vertical permeability distribution of the continental crust, i.e. exponentially decreasing with depth with an initial permeability $K_0 = 5.10^{-6} m.s^{-1}$, see section 2.4. Models differ by the characteristic depth value corresponding to $z_0 = 250 m$ for K_f , $z_1 = 125 m$ and $z_2 = 25 m$ for K_{f_1} and K_{f_2} , respectively. A lower characteristic depth leads to a decrease much stronger in permeability (Figure 45). Simulations for K_f, K_{f_1} (Figure 47) reflect the same trend than homogeneous models for a period of 100 years with a rapid increase followed by a stabilization. A decline then occurs on the long term due to the permeability distribution as shown in the case of K_{f_2} (Figure 47). According to plume evolution (Figure 45) for the three time steps (5 yr, 20 yr and 100 yr) this first stage seems to correspond to initial development of gravity instabilities until a more efficient configuration is reached. Figure 47 shows that permeability distribution affects normalized mass flux and consequently the evolution of average concentration of the considered domain. A higher decline in permeability results in a decrease of normalized mass flux and in lower average concentration which represents the cumulative effect of the process over time. When permeability becomes too low the density contrast is not sufficient to allow an efficient propagation of seawater at depth (Figure 46: K_{f_2}). When permeability decreases quickly, salinization process and extension of the plume are rapidly limited and normalized mass flux tends to decrease (clearly visible for K_{f_2} in Figure 47 and Figure 46). For K_{f_1} this process is less visible but initiated with a slightly decline of normalized mass flux.

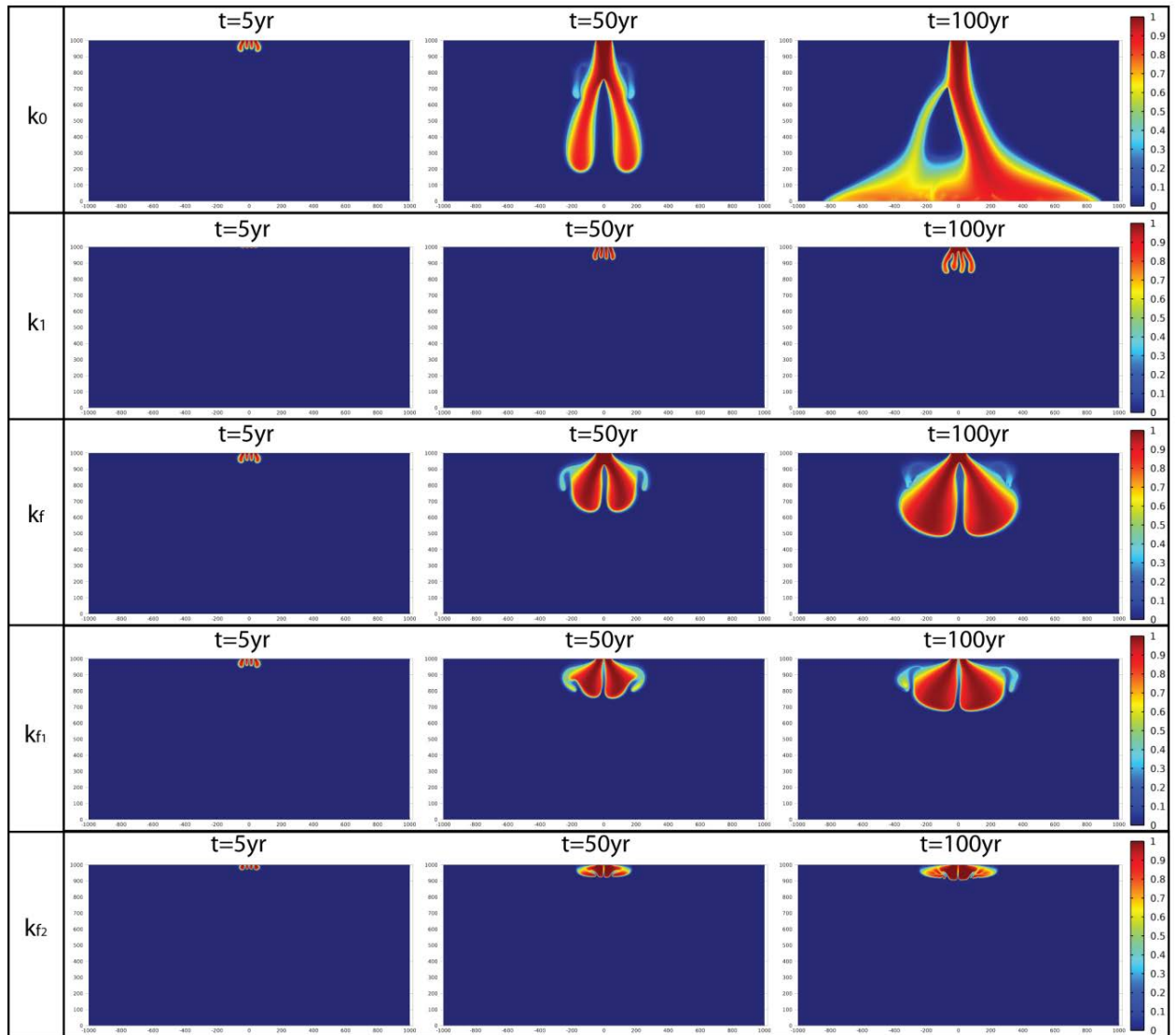


Figure 46. Evolution of gravity instabilities for 3 time steps (5yr, 50yr and 100yr) according to the 5 permeability models (K_0 , K_1 , K_f , K_{f1} and K_{f2}).

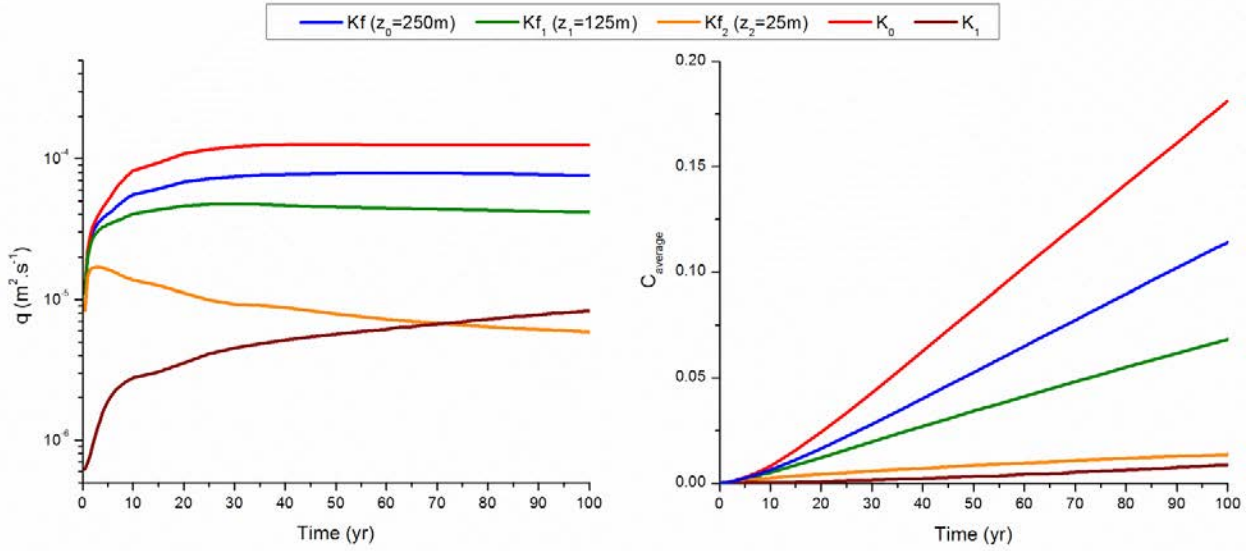


Figure 47. Normalized mass flux versus time for the 5 permeability modeled: Homogeneous (K_0 , K_1) and Exponential (K_f , K_{f_1} and K_{f_2}) distributions (left). Evolution of average concentration of the domain versus time for the 5 permeability modeled (right).

Results of normalized mass flux as function of properties of the groundwater system suggest a relationship between permeability and salinization efficiency. Normalized mass flux value for a time of 100 yr for each simulation was expressed as a function of the domain permeability (Figure 48). For exponential distribution models (K_f , K_{f_1} and K_{f_2}), the permeability corresponds to:

$$K_w = K_0 \cdot e^{-\frac{L_c}{z}}$$

where L_c is the critical river length over which salinity is efficient ($L_c < W$) and z is the characteristic depth value corresponding to $z_0 = 250 \text{ m}$ for K_f , $z_1 = 125 \text{ m}$ and $z_2 = 25 \text{ m}$ for K_{f_1} and K_{f_2} , respectively.

Figure 48 shows a linear relationship between salinity normalized mass flux and permeability of the groundwater system. Thus, normalized mass flux can be written as a function of the groundwater system permeability:

$$q_K^* \sim \frac{q}{W \cdot \frac{\Delta \rho}{\rho_0}} \sim f \cdot K_w$$

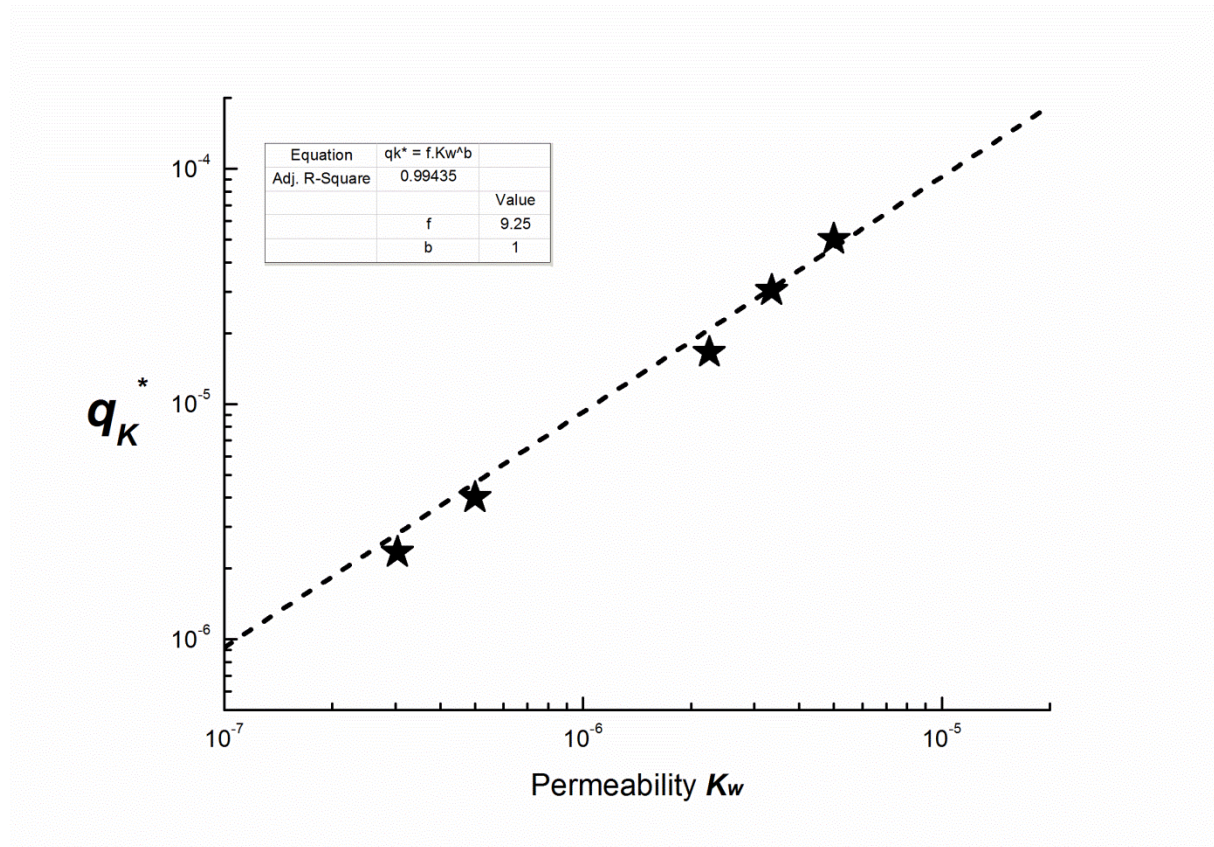


Figure 48. Inflow rate as function of average permeability of the domain considered

3.1.3. LONG TERM EVOLUTION OF GRAVITY INSTABILITIES

Long term simulations have been computed for the 3 exponential permeability distributions (K_f , K_{f_1} and K_{f_2}). In comparison with homogeneous models K_0 and K_1 , the time to reach the bottom is much longer because at 400 years gravity instabilities have travelled approximately 850 m and 500 m for K_f and K_{f_1} , respectively (see Figure 49). Moreover, for K_{f_2} instabilities are still in the “shallower” part (100m) (Figure 49). Prolongation of simulation on a longer term shows an interesting behavior characterized by a new phase of destabilization clearly visible for K_f and K_{f_1} , as shown in Figure 49.

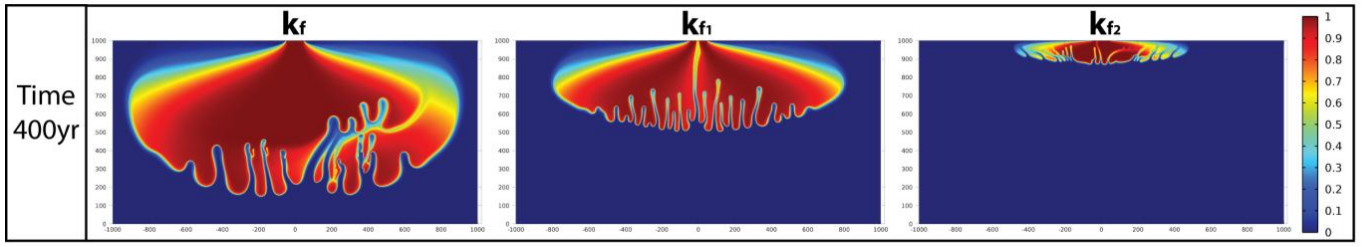


Figure 49. Gravity instabilities for a time period of 400 years according to the 3 models of permeability (K_f , K_{f1} and K_{f2}).

Evolution of plumes has been studied using horizontal and vertical dispersions of the plume over time for these permeability models (K_f , K_{f1} and K_{f2}) (Figure 50). Dispersions have been calculated for concentrations $C > 1e - 4$ in horizontal (x) and vertical (z) directions with:

$$x_{dispersion} = \sqrt{\frac{\sum_{i=1}^N C_i (x_i - \bar{x}^*)^2}{\frac{N' - 1}{N'} \sum_{i=1}^N C_i}}$$

$$z_{dispersion} = \sqrt{\frac{\sum_{i=1}^N C_i (z_i - \bar{z}^*)^2}{\frac{N' - 1}{N'} \sum_{i=1}^N C_i}}$$

where $\bar{x}^* = \frac{\sum_{i=1}^N C_i x_i}{\sum_{i=1}^N C_i}$ and $\bar{z}^* = \frac{\sum_{i=1}^N C_i z_i}{\sum_{i=1}^N C_i}$

Vertical and horizontal dispersions reflect the effect of permeability distributions, with an easily readable graduation visible on Figure 50 , showing higher values for K_f and lower values for K_{f2} . The most important point is the competition between vertical and horizontal dispersions over time. At the beginning for K_f vertical propagation of the plume is dominant but after a time of 200 years horizontal extension becomes dominant. On the contrary, for K_{f1} and K_{f2} horizontal extension is dominant much sooner (after 25

years in K_{f_1} and from the beginning in K_{f_2} , Figure 50). Moreover, the horizontal prevalence tends to increase over time in all cases. Limitation of the salinity propagation at depth promotes the horizontal extent of the plume due to the permeability distribution with depth. On the short term the horizontal prevalence promotes the destruction of fingers, bringing together fingers (K_f and K_{f_1} Figure 46) but on the long term it gives birth to a new spectrum of instabilities (K_f and K_{f_1} Figure 49). This behavior (preferential horizontal extension of the plume) is clearly in contrast with the vertical propagation for the homogenous domain (Figure 46 and Figure 49). The preferential horizontal extension is the result of the exponential permeability distribution and more specifically of the contrast between the horizontal and the vertical permeability where $K_L > K_v$.

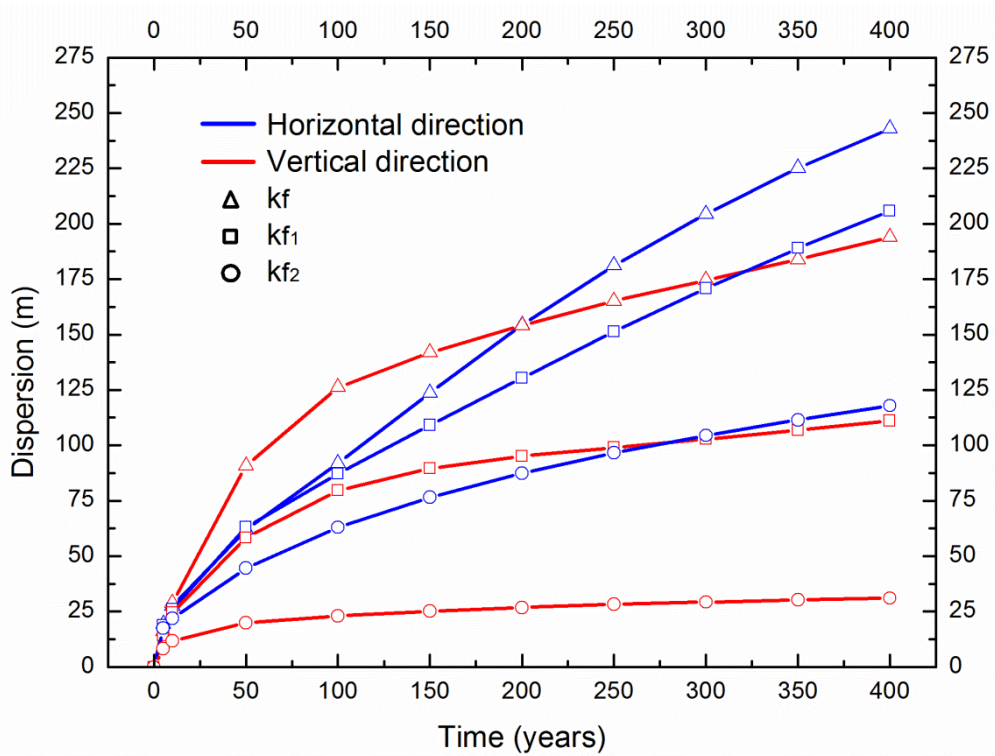


Figure 50. Plume evolution in x-direction and y-direction over time for the three exponential permeability models (K_f , K_{f_1} and K_{f_2}).

3.2. NORMALIZED MASS FLUX AS A FUNCTION OF RIVER CONCENTRATION

As stated previously, the leading edge of seawater entering the fluvial system due to connection with the sea and river salinity evolves with the distance from the sea (Figure 42). At the estuary the water density is around 1025 kg.m^{-3} (seawater density, $\frac{C_{river}}{C_s} = 1$) and evolves until the freshwater density 1000 kg.m^{-3} at several kilometers from the sea. To quantify the effect of river concentration on normalized mass flux, simulations were done with a range of concentration at the river boundary (Table 1). Normalized mass flux over time was calculated for each concentration and plotted as function of time (see Figure 51).

First, all simulations reflect the same trend for a period of 100 years with a rapid increase followed successively by a stabilization as described in the previous section (Figure 51). Time required to reach the maximum normalized mass flux is dependent on the concentration. Indeed, this time increases with the reduction of the density contrast so with the velocity reduction. Variations observed due to difference of concentration at the river boundary are consistent with instabilities propagation (Figure 52). For the highest concentration ($\frac{C_{river}}{C_s} = 1$) and so for the highest density contrast depth reached by instabilities is higher due to the greater velocity which is controlled by the density contrast.

Evolutions of normalized mass flux as a function of concentration suggest there is a relationship between these parameters (Figure 51). The mean normalized mass flux normalized for a time of 100 yr for each simulation was plotted as function of the concentration applied at the river boundary (Figure 53). This log-log graph demonstrates clearly a relationship between river concentration and normalized mass flux. The fitted curve indicates that normalized mass flux evolves as function of the concentration square:

$$q_c^* \sim \frac{q}{W \cdot \frac{\Delta\rho}{\rho_0} \cdot K_w} \sim f \cdot C^2$$

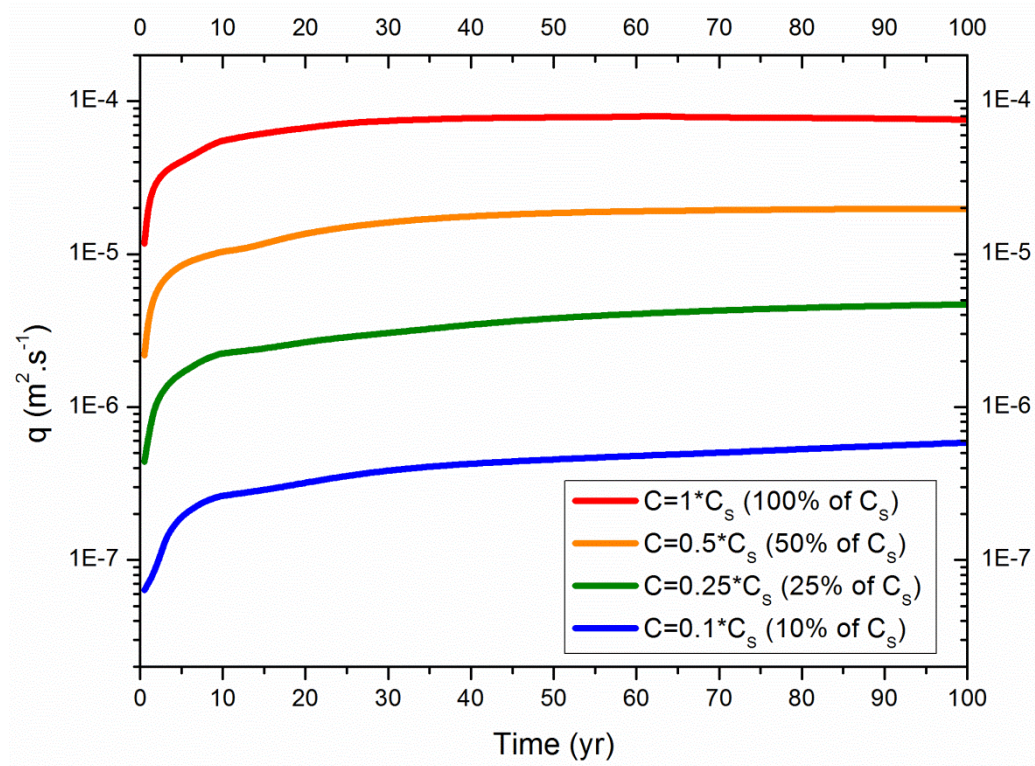


Figure 51. Inflow rate versus time for 4 concentrations at the river boundary (percentage of seawater concentration: 100, 50, 25 and 10%).

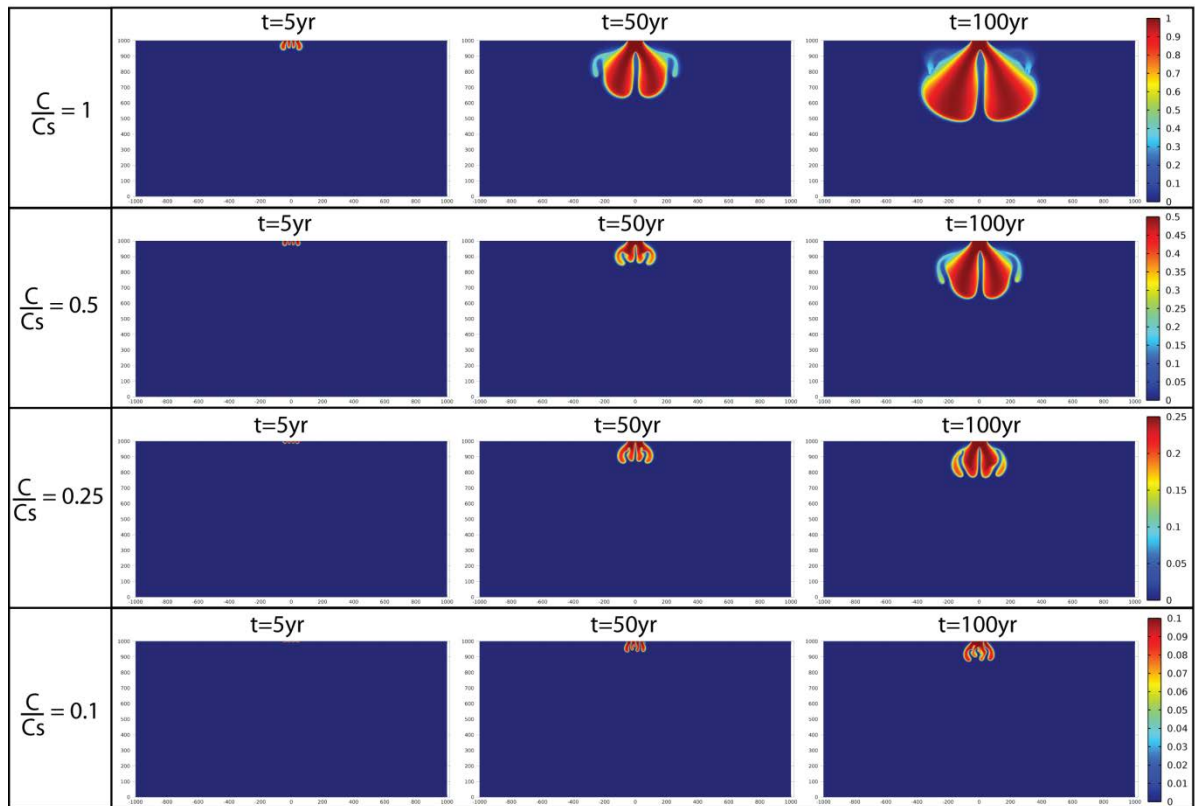


Figure 52. Evolution of gravity instabilities for 3 time steps (5yr, 20yr and 100yr) according to 4 concentrations at the river boundary (percentage of seawater concentration: 100, 50, 25 and 10%).

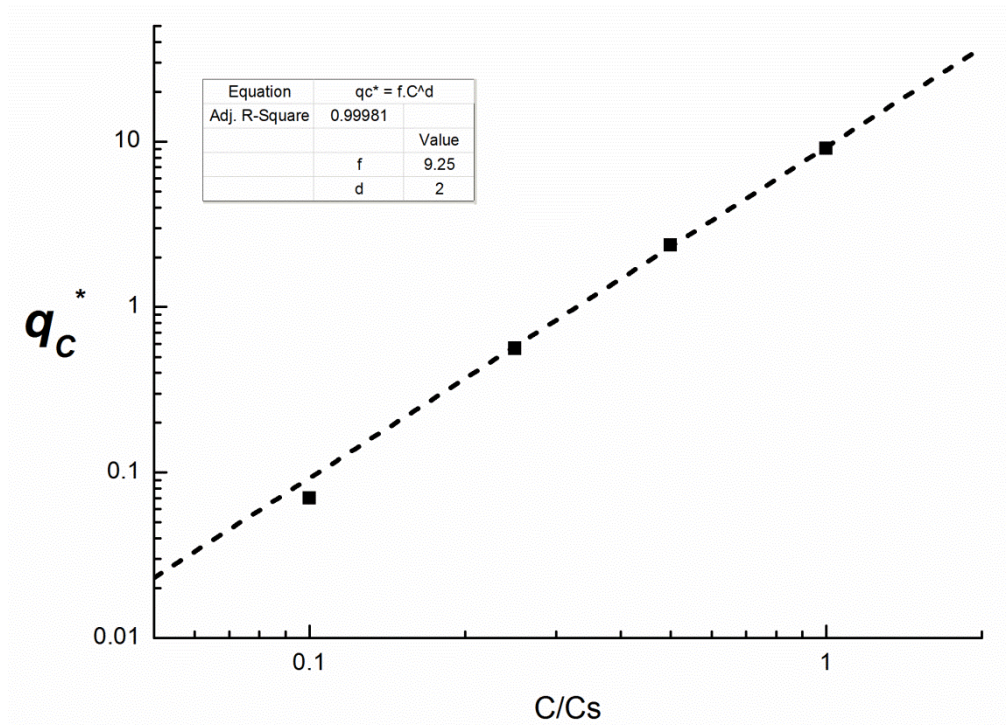


Figure 53. Inflow rate as function of concentration

3.3. NORMALIZED MASS FLUX AS FUNCTION OF RIVER WIDTH

Natural fluvial systems are typically characterized by wide ranges in river width (W). Thereby variations in the width of rivers may induce a wide range of normalized mass fluxes. To investigate the effect of channel width on salinity normalized mass flux, simulations were done with a range of river width which varies on three orders of magnitude (Table 1). normalized mass flux over time has been calculated for each width and plotted as a function of time (Figure 54). Two different behaviors can be observed. For river width ranging from 100 to 1m the same trend (as observed previously) is visible with a rapid increase of normalized mass flux followed by stabilization. However, for rivers wider than 100m, the behavior seems to be more unstable with fluctuations over time (Figure 54). Nevertheless, normalized mass fluxes appear to fluctuate around an average value. Unstable behaviors of river widths of 1000m and 500m are visible in Figure 55 showing gravity instabilities development over time. Results clearly show various evolutions of gravity instabilities through time for each order of magnitude (Figure 55). Despite differences, normalized mass flux is clearly dependent on river width with increasing fluxes when the river width is larger. However, it seems that salinization efficiency is limited for a small river width. Indeed for rivers of 10 and 1m,

only one plume has developed the front concentration tends to decrease with depth and normalized mass flux is reduced.

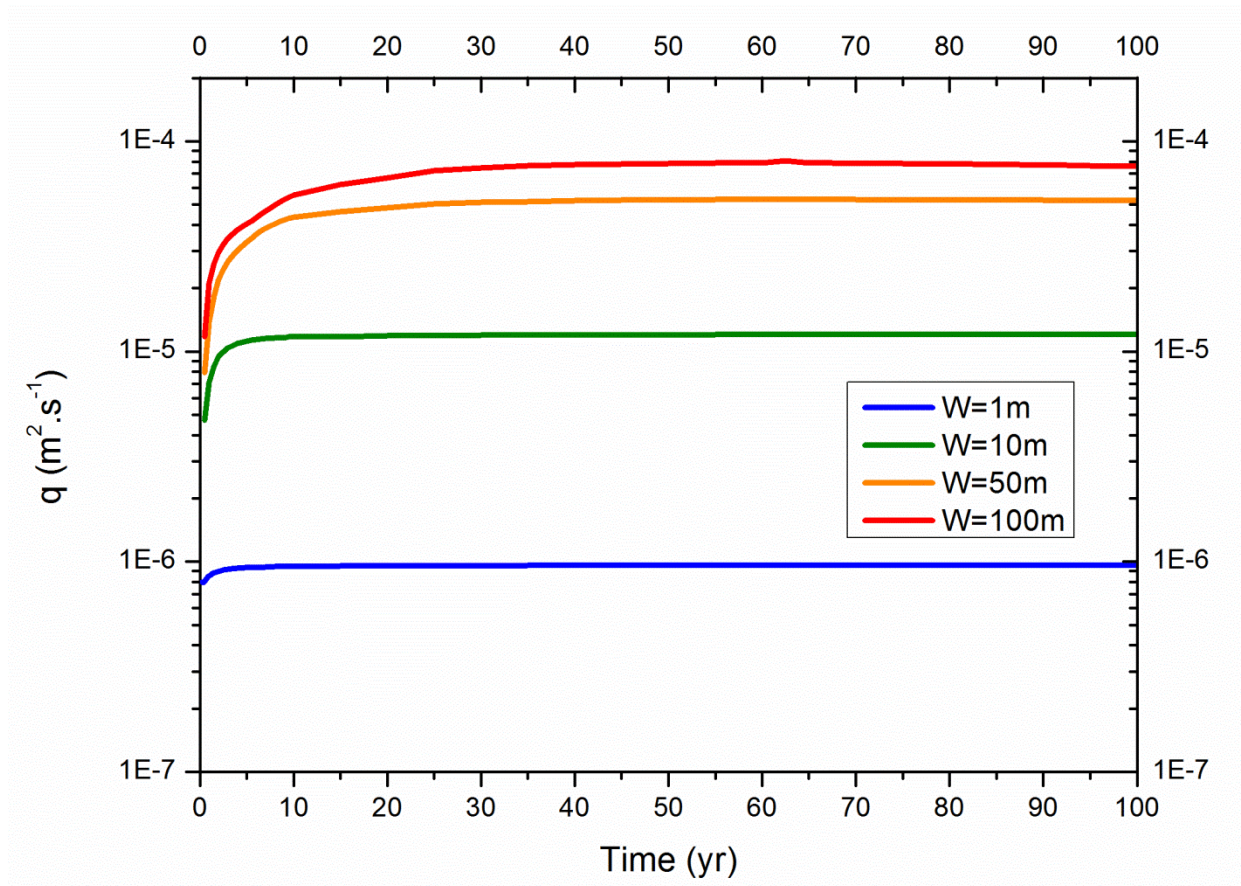


Figure 54. Normalized mass flux vs time for 5 river widths (1, 10, 50 and 100 m)

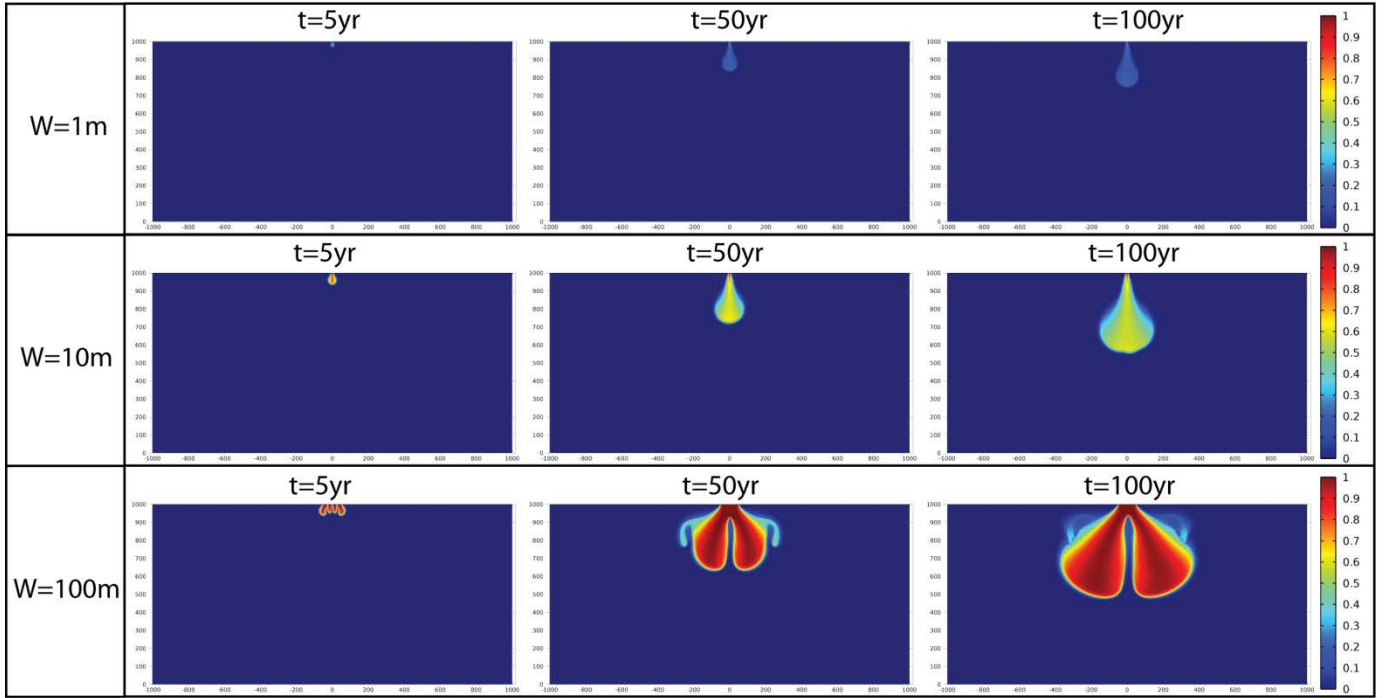


Figure 55. Evolution of gravity instabilities for 3 time steps (2.5yr, 20yr and 100yr) according to 3 river widths on three orders of magnitude (1, 10 and 100 m).

Mean normalized mass flux normalized by the river width, the permeability and the density contrast for a time of 100 yr for each simulation has been plotted as a function of river width (Figure 56). These results enable to calculate the parameter f (estimated to approximately $f \sim 9.25 \pm 0.85$ in this study) below which the normalized mass flux remains limited:

$$q_w^* \sim \frac{q}{W \cdot K_w \cdot \frac{\Delta \rho}{\rho_0}} \sim f$$

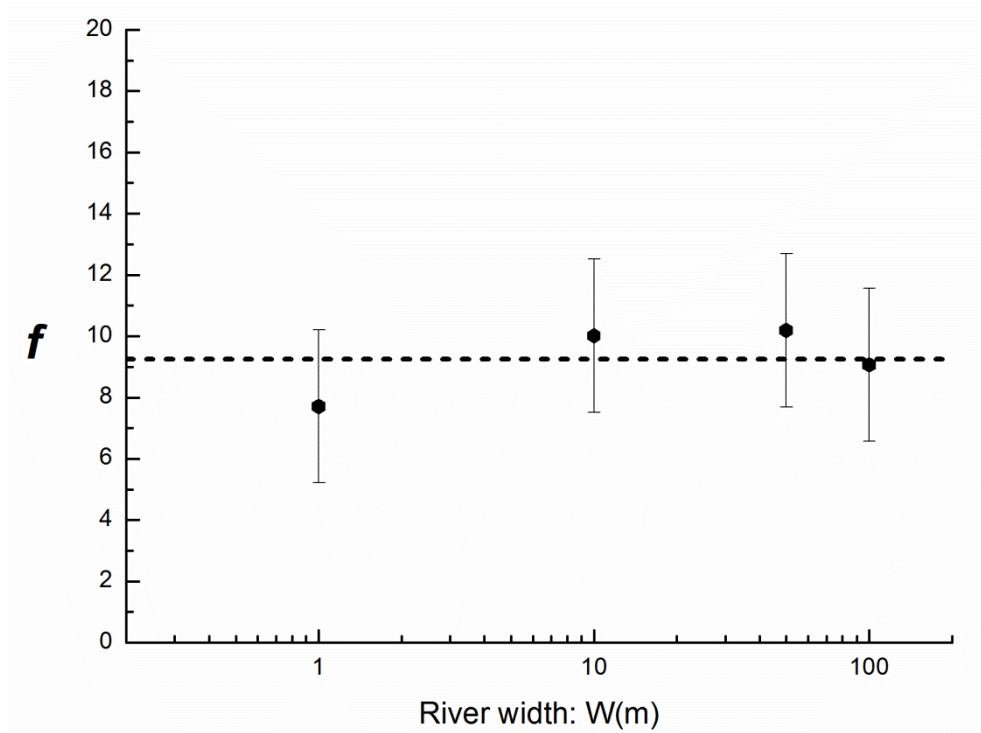


Figure 56. Normalized mass flux as function of river width

3.4. NORMALIZED MASS FLUX GENERALIZATION

Considering the three relationships established previously between salinity normalized mass flux and the groundwater system permeability, the river width and the river concentration, we obtain:

$$q_K^* \sim \frac{q}{f \cdot K_w \cdot W \cdot \frac{\Delta \rho}{\rho_0}}$$

$$q_C^* \sim \frac{q}{f \cdot C^2 \cdot W \cdot \frac{\Delta \rho}{\rho_0} \cdot K_w}$$

$$q_W^* \sim \frac{q}{f \cdot W \cdot K_w \cdot \frac{\Delta \rho}{\rho_0}}$$

Thus, normalized mass flux Q [$\text{m}^2.\text{s}^{-1}$] corresponding to an equivalent volume of seawater per unit of time and of river width can be generalized, as:

$$Q = f \cdot K_w \cdot W \cdot C^2 \cdot \frac{\Delta\rho}{\rho_0}$$

where $f \sim 9.25$

Considering the salinity of seawater ($C_s = 35\text{kg}.m^{-3}$), the quantity of salinity (Q_s) [$\text{kg}.\text{s}^{-1}$] per unit of time can be obtained from Q , as:

$$Q_s = C_s \cdot f \cdot K_w \cdot W \cdot C^2 \cdot \frac{\Delta\rho}{\rho_0}$$

$$Q_s = 35 \cdot f \cdot K_w \cdot W \cdot C^2 \cdot \frac{\Delta\rho}{\rho_0}$$

On the other hand, salinity evolves with the distance from the sea and is well fitted by an exponential curve (Figure 42):

$$C(x) = C_s \cdot \exp^{-\frac{x}{x_0}}$$

where C_s the seawater concentration and x_0 the characteristic distance along which the leading edge of seawater entering the fluvial system is significant. Inflow rate is dependent on the concentration square where concentration is a function of the distance from the sea:

$$\int_0^X \left(\frac{C(x)}{C_s}\right)^2 dx = \int_0^X \left(\exp^{-\frac{x}{x_0}}\right)^2 dx = \left[-\frac{x_0}{2} \cdot \exp^{-\frac{2x}{x_0}}\right]_0^X = \frac{x_0}{2} \left(1 - \exp^{-\frac{2X}{x_0}}\right) \text{ with } X \rightarrow +\infty$$

Thus, the salinization process is important over half of the characteristic length $\frac{x_0}{2}$. Assuming that the significant distance of each river can be approximated by:

$$x_0 \sim \frac{\Delta h_{sea}}{S}$$

where S is the river slope and Δh_{sea} the average tidal range salinity inflow rate (Q_s) [$\text{kg}.\text{s}^{-1}$] along one river is:

$$Q_s = 35. f. K_w. W. \frac{\Delta \rho}{\rho_0} \cdot \frac{\Delta h_{sea}}{2. S}$$

To finish, global salinity fluxes at large scale will be function of the number of river (N_W) with a width W_i :

$$Q_{sTot} = \sum_{i=W_{min}}^{i=W_{max}} N_{W_i} \cdot Q_s(W_i)$$

3.5. NORMALIZED MASS FLUX VS UPWARD GROUNDWATER FLOW

As specified previously, streams receive water from groundwater discharges occurring in horizontal and vertical directions to the stream and thus in opposition with the propagation of instabilities. To investigate the effect of upward flow on salinity inflow rate, simulations were done with fresh groundwater flow which feeds fluvial system. The top boundary is modeled with a hydraulic gradient ($h=i$) on each side of the river and a fixed head ($h=0m$) is applied to the river. A zero concentration value ($C=0$) is applied to the top boundary of each side of the river.

The investigation of the critical hydraulic gradient has been realized with a river concentration ($\frac{C_{river}}{C_s} = 1$), a river width of 100m and a homogeneous system permeability (K_0). The critical hydraulic gradient (i_c) corresponds to the maximum value, above which the salinization process is inefficient. Here, for the specified case ($\frac{C_{river}}{C_s} = 1$; $W = 100m$ and $K = K_0$), the critical hydraulic gradient is $i_c = 0.0037$ as shown by results with $i_c > 0.0037$ instability does not exist and salinity inflow rate is null (Figure 57: $i_c = 0.0038$) in contrast to situations where $i_c \leq 0.0037$ (Figure 57: $i_c = 0.0037$ and 0.0036). Salinity inflow rate is proportionally limited to upward groundwater flow, indeed when the hydraulic gradient is higher the gravity instabilities propagation is slightly restrained (Figure 57: $i_c = 0.0037$ and 0.0036).

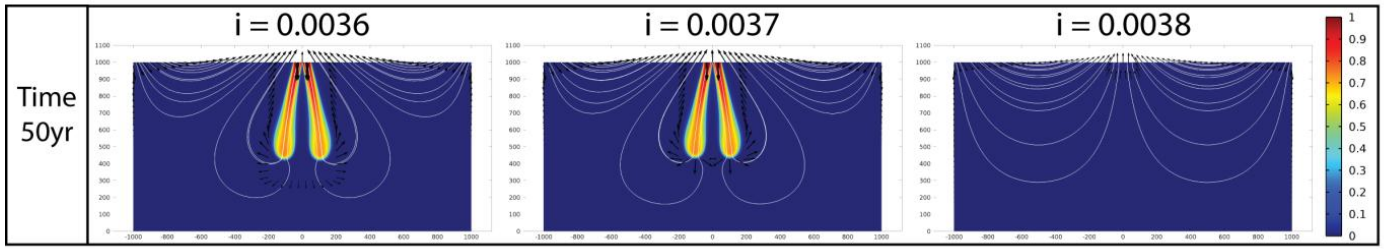


Figure 57. Gravity instabilities in function of upward groundwater flow for three hydraulic gradients (0.0036, 0.0037 and 0.0038) for a time step of 50 years. White line represents streamlines according to horizontal and vertical velocity field. Black arrows show the groundwater flow directions.

4. DISCUSSION

The main aim of this study was to characterize groundwater salinization efficiency below coastal rivers. Understanding and quantifying the groundwater salinization process are important because it provides significant constraints on the quantity of seawater that may enter into a fresh groundwater system due to salinized rivers. Using numerical experiments, we examined salinization efficiency according to three major parameters which characterize the salinization concept (Figure 43): the channel width, the density contrast (between salinity degree of the river and fresh groundwater) and the groundwater system permeability. It is suggested that numerical experiments carried out in this study have a generic value in the sense that the behavior tested in each experiment is representative of global behaviors (in term of processes). However, absolute values will depend on the system geometry and on the permeability distribution. This assumption is based on the fact that the various existing situations found in nature (e.g. river properties, salt front evolution, groundwater system permeability...) are taken into account by the different parameters tested in numerical experiments. This idea can be easily understood from the conceptual model (Figure 43). Although we have not systematically varied all system parameters (e.g. porosity, horizontal and vertical dispersion, aquifer dimension...) and that natural settings may be more complex, experiments computed are consistent with this hypothesis.

The porosity effects have been reported in the case of study on CO₂ storage in saline aquifers (Pau et al., 2010). A decrease in porosity (θ) increases the effect of the convective process and fingers become thinner. In this study, we performed simulations with a porosity of 10 %, but it is almost certain that the real porosity in continental

aquifers is lower and suggests a stronger inflow rate. Similarly, the effects of dispersion on fingering velocity have been previously studied (Xie et al., 2011). An increase in longitudinal dispersion (α_L) increases the dissipation of the solute and thus reduces the velocity. A decrease in vertical dispersion (α_T) provides thinner fingers and reduces the velocity, probably due to a smaller density contrast.

However, in the most natural cases, streams receive water from groundwater discharges which occur in horizontal and vertical directions to the stream and hence are in opposition with the direction of free convection process. Indeed, as stated previously, quantity introduced is limited by fresh groundwater flow which feeds streams. Results suggest that salinization process is inefficient when:

$$V_g \leq V_f$$

$$f_c \cdot \frac{\Delta}{0} \leq i_c$$

This above condition suggests that groundwater systems which are influenced by gravity instability below salinized rivers are dependent on the salinity degree of the river and the hydraulic gradient and therefore directly correlated to the topography close to the river. That's why a simple analysis of the topography (topography slope transversal to the river) of the domain considered will enable to produce regional cartography of area influenced by this salinization mechanism. For example, considering the maximum density contrast (which corresponds to salinized river next to the sea) a regional slope of the order of 1.25 % is required to limit the salinization process. Nevertheless, as shown by results in section 3.5, the corrective factor is not constant and changes with the system permeability. Thus, it is necessary to take into account the corrective factor variability. Moreover seawater inflow rate is limited proportionally to the groundwater fluxes and the salinity patterns is affected. This natural factor requires further investigation. However, as a first step, it seems reasonable to consider that this study provides a reasonable estimation of groundwater salinization below rivers influenced by seawater in low-lying areas with limited topography on each side of a coastal river.

The general formula provides a framework for estimation of current salinity fluxes into fresh groundwater at large scale. In fact according to characteristic properties of natural systems (salinity profile, river width, drainage basin area...) an overview of salinization inflow rate into fresh groundwater at large scale may be produced. Geomorphological studies have described a number of relationships to quantify processes occurring at the earth surface according to characteristic features of landscape. Consequently channel width (W) and local slope in bedrock catchment have been linked with drainage area (A) through relationships, where $S \sim A^\alpha$ and $W \sim A$ (Hack, 1957; Montgomery and Gran, 2001). From a network of rivers at the regional scale, the drainage basin areas associated to rivers connected to the sea can be identified and calculated. On the basis of a classification of the drainage basin area, an average local slope can be determined for each range according to the study of area-slope relationships ($S \sim A^\alpha$) (Lague et al., 2000). Similarly, average river width (next to the estuary) for each range can be estimated from the area-channel width relationships ($W \sim A$). Inflow rate q_s for each river width can be calculated as a function of drainage basin area and considering an average tidal range characteristic of the study region. Then, regional salinity inflow rate can be calculated from the general formula taking account the number of river with a width W_i and where the salinization process is efficient. Thus, the quantification will be realized in agreement with a risk map previously edited and directly established from the topography close to the river.

Because the ubiquity of rivers connected to the sea, it seems that salinization process of continental aquifers due to salinized rivers may be a quite common process and need to be more investigated. Moreover, in the context of climate change which is expected to lead to sea-level rise and consequently the progression of saline water inland, aquifers salinization below coastal rivers could be more significant.

5. CONCLUSION

Seawater intrusion in continental aquifers by gravity instabilities below coastal rivers was investigated. This study is motivated by the fact that salinized rivers occur worldwide and may widely contribute to groundwater salinization by the setting of seawater over fresh groundwater systems. Here, we demonstrated one of the most efficient processes in macrotidal environments, the gravity-driven instability that induces downward salt water invasion below coastal rivers. Using numerical experiments salinity inflow rate was investigated as a function of three characteristic parameters to be consistent with natural settings of these systems. Results provide a general formula to quantify salinity inflow rate according to four main parameters: (1) the groundwater system permeability, (2) the river width and its slope, (3) the salinity of the river and (4) the tidal amplitude. The findings provide a framework for the prediction of groundwater salinization at large scale under present-day conditions. The general expression was used to estimate salinization rates in Brittany, given the geomorphological and environmental characteristics (drainage basin area, river widths and slopes, tidal range, aquifer permeability). We found that invasion below coastal rivers entails very high saline water inflow rates, indicating that this process may play a major role in the salinization of regional aquifers. Moreover, the findings of this study could be applied in the context of climate change where a sea-level rise is expected in order to provide an estimation of the groundwater salinity evolution. This last question constitutes a major issue for the management of groundwater resources in the near future. The next steps of this study will be to produce a current mapping of seawater invasion at the regional scale, as well as a possible mapping considering the future according to climate change and provides an estimation of the potential volume of fresh groundwater affected.

Acknowledgments:

Data supporting Figure 1 is available at <http://www.loire-estuaire.org/dif/do/init>.

4.4. Conclusion

Cette étude se focalise sur la problématique des processus de salinisation des aquifères. À ce jour, les processus de salinisation ont principalement été étudiés dans le contexte des aquifères côtiers, néanmoins la salinisation des aquifères à travers le système fluvial n'a pas été prise en compte. Or, les rivières sont en interaction à la fois avec les aquifères et l'océan.

À l'heure actuelle, l'influence de l'océan se fait déjà fortement ressentir sur la salinité des eaux de rivière à des distances plurikilométriques à l'intérieur des terres. Cette incursion d'eau de mer dans le système fluvial entraîne la mise en place d'une stratification densitaire avec de l'eau de mer au-dessus d'aquifères d'eau douce. Sous l'effet de cette différence de densité, entre l'eau salée au sein du réseau hydrographique et de l'eau douce dans la nappe souterraine la salinisation des aquifères peut avoir lieu.

L'étude présentée propose une quantification de ces processus de salinisation en fonction de paramètres caractéristiques du milieu naturel : la largeur de la rivière, la proportion d'eau de mer dans la rivière et la perméabilité du milieu souterrain sous-jacent. Les résultats de ces simulations permettent d'établir une loi permettant de quantifier la salinisation en fonction de ces 3 paramètres. Une application en est faite à l'échelle de la Bretagne en tenant compte de différentes caractéristiques (largeur des rivières, nombre et pente des rivières, hauteur moyenne du marnage, perméabilité moyenne) dans le but d'apprécier l'impact de la salinisation des aquifères par le réseau hydrographique à grande échelle.

Cette application semble mettre en avant le fait qu'à l'échelle régionale les petites rivières participent légèrement plus à la salinisation des aquifères que les grandes du fait de leur plus grand nombre. Néanmoins, les pentes de ces petites rivières étant plus fortes, l'introduction de l'eau de mer vers l'intérieur des terres est plus limitée en distance alors que dans les grandes rivières le front de salinité progresse plus loin et entraîne une salinisation des aquifères à une distance plus éloignée des côtes. Enfin, la quantification à l'échelle globale annonce un volume important d'eau de mer pouvant être introduit par ce processus, soulignant ainsi l'importance de considérer le réseau fluvial comme un acteur majeur dans la salinisation des aquifères.

Chapitre 4. Relation eau souterraine – eau de mer et climat

Par conséquent, il convient de s'intéresser plus amplement à ce processus de salinisation puisque dans les conditions actuelles (climat, niveau de la mer) les rivières semblent être d'une importance majeure. Or, dans un contexte de changement climatique, le niveau de la mer devrait augmenter et amplifier le phénomène de la salinisation des aquifères par les rivières, du fait de la progression du front de salinité à l'intérieure de celles-ci.

Chapitre 5. Indices paléoclimatiques dans les eaux souterraines: Apport de la modélisation numérique à densité variable

5.1. Introduction

Les fluides salés dans les aquifères de socle ont été répertoriés à diverses profondeurs et à travers le monde entier, témoignant de leur omniprésence (Bucher and Stober, 2010; Frapet et al., 2003). L'origine de ces fluides est encore à ce jour un sujet de débat en particulier à cause des fortes interactions entre l'eau et la roche encaissante amenant à la modification de la signature géochimique originelle de ces eaux. Néanmoins, dans plusieurs cas, l'origine de ces fluides a pu être attribuée à de l'eau de mer ancienne qui se serait introduite dans les aquifères de socle au cours de transgressions marines passées (Aquilina and Dreuz, 2011; Armandine Les Landes et al., 2014; Beaucaire et al., 1999; Bottomley et al., 1994; Cloutier et al., 2009; Greene et al., 2008; Louvat et al., 1999). Bien que les mécanismes d'introduction d'eau de mer et les constantes de temps associées restent encore mal compris, des indices de plus en plus nombreux soulignent l'existence de grande circulation d'eau d'origine marine en profondeur des systèmes souterrains.

Au sein du Massif Armorica, des fluides salés possédant des concentrations modérées ont été échantillonnés au sein des aquifères de socle et analysés à l'échelle régionale (Armandine Les Landes et al., 2014; Ayraud et al., 2008; Pauwels et al., 2010). La récente analyse de la distribution horizontale et verticale à l'échelle régionale des hautes concentrations en chlorure (60-1400mg/L) a révélé l'origine marine de ces fluides. En effet, les distributions des paléocôtes relatives aux transgressions du

Pliocène, mettent en évidence la bonne corrélation entre les concentrations anormales en chlorure et les zones inondées durant les transgressions. Une remarque particulière peut être apportée vis-à-vis de ces résultats. Cette étude s'appuie en partie sur de précédentes études (géologiques, stratigraphiques, paléontologiques...) utilisant une courbe eustatique avec des amplitudes qui ont depuis été révisées. Néanmoins, des travaux de thèse actuellement réalisés au sein de Géosciences Rennes ayant pour sujet : l'évolution géomorphologique du Massif armoricain depuis 200 Ma (Bessin, 2014) témoigne de la bonne adéquation des résultats tout en considérant les nouvelles courbes eustatiques édifiées. Ces informations apportent des contraintes précises sur l'âge de ces fluides ainsi que sur l'histoire du système hydrogéologique (Armandine Les Landes et al., 2014).

En effet, durant une transgression marine, de l'eau de mer s'est introduite à grande profondeur dans le socle (écoulement conduit par effet densitaire) ainsi que profondément à l'intérieur des terres. Suite à la phase de transgression, le cycle hydrologique continental ainsi que les écoulements naturels souterrains se sont remis en place. Sous l'effet de ces écoulements souterrains alimentés par les eaux météoriques, le reliquat d'eau de mer est progressivement lessivé. La distribution actuelle des eaux souterraines, à la fois douce et salée, au sein des aquifères est donc principalement le résultat de processus prenant place sur des temps géologiques (processus naturels d'intrusion d'eau de mer suivis par un processus de lessivage d'eau souterraine possédant une origine météorique). D'autre part, les activités anthropiques ont pu aussi avoir eu un effet sur ce cycle, néanmoins celles-ci se font dans des proportions plus limitées en termes d'échelle de temps et de distance. Cette signature géochimique marine identifiée au sein d'une majeure partie de la Bretagne souligne que les processus d'intrusion d'eau de mer ne sont pas restreints aux aquifères côtiers mais permettent aussi l'introduction d'eau de mer jusqu'à l'intérieur des terres. Ces indices géochimiques nous ont donc conduits à nous intéresser aux mécanismes d'intrusion d'eau de mer dans les aquifères continentaux dans le but de comprendre l'introduction « irréversible » d'eau de mer dans le socle.

C'est dans cette optique que la modélisation des écoulements à densité variable a été initiée. La modélisation numérique réalisée à partir du logiciel COMSOL Multiphysics a donc permis d'étudier ces processus de salinisation des aquifères dans le but de répondre à deux questions principales sous-jacentes qui sont d'une part la salinisation passée des aquifères durant une phase de transgression marine et d'autre part l'apport d'informations sur la dégradation possible de la qualité des eaux souterraines dans le cadre du changement climatique et de l'augmentation du niveau de la mer attendue.

Or, dans le cadre de la problématique des fluides profonds salés et de la mise en évidence de l'importance de l'effet densitaire dans la conservation de la signature marine (évolution de la concentration avec la profondeur), la modélisation des écoulements à densité variable peut encore être utilisée. En effet, dans le but de compléter l'histoire des chlorures, il reste à comprendre l'évolution de la signature marine sous l'effet des écoulements après son introduction dans le système et plus particulièrement le processus de lessivage.

5.2. *Chlorures: perspectives de modélisation numérique*

Les informations paléohydrogéologiques, la conceptualisation du fonctionnement hydrogéologique du système ainsi que les caractéristiques qualitatives (géochimie, etc..) et quantitative (piézométrie...) de l'état actuel du système ouvrent de nombreuses perspectives en terme de connaissance du fonctionnement des aquifères en domaine de socle à l'échelle régionale. En effet, toutes ces informations constituent un solide point de départ pour de futurs travaux de modélisation numérique des écoulements de ces milieux à grande échelle de temps et d'espace. Il peut être envisagé de réaliser des simulations dans le but de raffiner et de corriger la compréhension conceptuelle du fonctionnement hydrodynamique. Depuis un certain nombre d'années, le développement de codes numériques capable de quantifier et d'évaluer l'évolution chimique des eaux a grandement progressé, donnant ainsi la possibilité d'interpréter les données géochimiques (Glynn and Plummer, 2005). Ce genre d'étude pourrait donc être mené au sein du Massif Armoricaïn en essayant par exemple de retracer l'histoire des chlorures d'origine marine.

Considérant la distribution verticale des concentrations en chlorure actuellement observée à l'échelle régionale et en posant certaines hypothèses sur l'état initial de la composition des eaux souterraines après une transgression marine. Des simulations d'écoulements et de transport (à densité variable) du lessivage de cette signature marine appliquées à la situation du Massif Armoricaïn pourraient apporter de nombreuses réponses et/ou contraintes sur les caractéristiques hydrauliques, géologiques ainsi que sur l'hydrodynamique de ces aquifères.

Dans cette optique, quelques travaux préliminaires en termes de modélisation ont été menés dans le but d'initier la compréhension de cette distribution verticale des chlorures. Dans un premier temps, l'hypothèse faite est que suite à une transgression marine, de l'eau de mer s'est introduite à grande profondeur dans le socle (processus conduit par effet densitaire). Suite à la transgression, le système de circulation à l'échelle régionale alimentée par des eaux météoriques se remet en place, donnant lieu au lessivage de cette empreinte marine. Ces modélisations ont été réalisées avec le logiciel COMSOL Multiphysics et simulent les écoulements à densité variable (Section 4.2 du chapitre précédent).

5.3. Conditions limites et initiales du modèle

Le système d'écoulements à grande échelle est représenté par un aquifère rectangulaire de 50 km de longueur et d'une épaisseur de l'ordre du kilomètre (de 1 à 4km). La limite inférieure et les limites verticales correspondent à des limites à flux nuls tandis que la limite supérieure correspond à une charge hydraulique $H(x)$ représentée par une fonction avec un terme sinusoïdale et un linéaire (pente régionale) qui induit les écoulements au sein du système (Toth, 1962). Une concentration constante nulle est appliquée à la limite supérieure (recharge par eau météorique) tandis que l'eau de mer est représentée par une concentration constante le long de la limite inférieure (Figure 58). Les paramètres utilisés dans ces simulations sont synthétisés dans le Tableau 6.

Chapitre 5. Indices paléoclimatiques dans les eaux souterraines: Apport de la modélisation numérique à densité variable

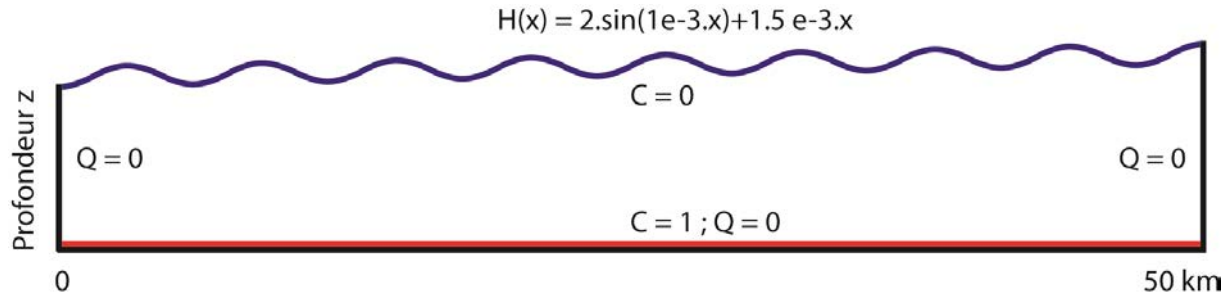


Figure 58. Conditions limites du modèle

Tableau 6. Paramètres de la modélisation

Symbole	Paramètre	Valeur	Unité
Dm	Coefficient de diffusion moléculaire	2.000E-08	m ² /s
g	Accélération de la pesanteur	9.81	m/s ²
Kf	Conductivité hydraulique	K=f(z)	m/s
θ	Porosité	0.1	
α	Coefficient de dispersivité longitudinale et transversale	20 ; 2	m
0	Densité eau douce (Référence)	1000	kg/m ³
S	Densité eau de mer	1025	kg/m ³
μ	Viscosité dynamique	1.00E-03	kg/m/s
z	Profondeur du domaine	1 à 4	km

5.4. Simulations et résultats

Ces premières modélisations ont été réalisées en régime permanent dans le but d'observer l'effet de ce type de système de circulation à l'échelle régionale comprenant des systèmes de différents ordres (locaux, intermédiaires et régionaux) sur une phase d'eau de mer introduite et située en profondeur du système. Les simulations ont été réalisées pour plusieurs profondeurs allant de 1 à 4 km.

La Figure 59 présente les écoulements simulés pour un système de 2 km de profondeur avec une conductivité hydraulique qui décroît avec la profondeur (Saar and Manga, 2004) (K_f section 2.4). Les lignes d'écoulement ainsi que les vitesses soulignent l'existence de systèmes de différents ordres, avec des écoulements plus rapides et plus courts dans la partie supérieure que dans la partie profonde. Ce partitionnement des écoulements est en accord avec les précédents travaux de modélisation réalisés par

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Goderniaux et al., (2013). Néanmoins, l'objectif premier ici est d'essayer de montrer l'influence de ce type d'écoulements sur la distribution des chlorures.

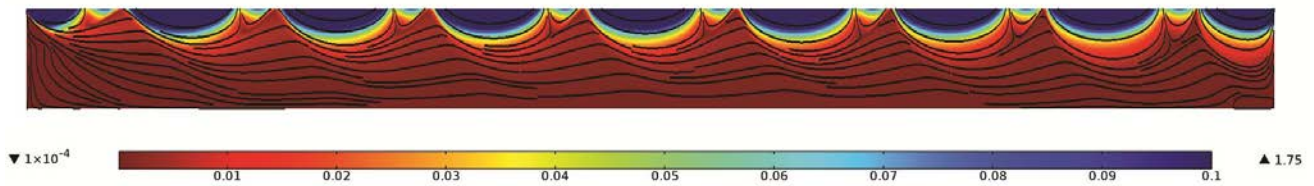


Figure 59. Organisation des écoulements simulée pour un système de 50km de longueur et de 2km de profondeur et avec une conductivité hydraulique exponentiellement décroissante avec la profondeur. Les résultats présentent l'organisation des écoulements (systèmes de différents ordres) par la présentation des vitesses de circulations (m/an) et des lignes d'écoulements correspondantes.

La Figure 60 présente les résultats des simulations en régime permanent réalisés pour un système de différente profondeur (1 à 4 km). Ces résultats semblent mettre en évidence l'effet du partitionnement des écoulements et plus particulièrement l'effet des zones de circulation active dans la partie supérieure avec un lessivage important, phénomène identifiable par la seule présence d'eau douce dans ces boucles.

Ces résultats peuvent aussi être présentés sous forme d'un profil vertical de la concentration moyenne du système. La Figure 61 présente ces profils et montre logiquement l'augmentation de la concentration avec la profondeur. Par contre, bien que l'ordre de grandeur des concentrations soit différent, les tendances de ces profils semblent présenter quelques similitudes avec la distribution des chlorures observée au sein des aquifères du Massif Armoricaïn à l'échelle régionale ainsi qu'avec la tendance générale présentée dans le Chapitre 2.2.

Cette tendance générale se caractérise par deux tendances distinctes entre la partie supérieure et la partie plus profonde avec entre les deux une zone de transition. Les pentes associées à ces zones peuvent être le résultat de processus particuliers mis en jeu dans le lessivage. Processus qui pourraient être caractérisés par une étude plus approfondie (identification des pentes caractéristiques, loi d'échelle, processus dominants en fonction de la profondeur, etc.).

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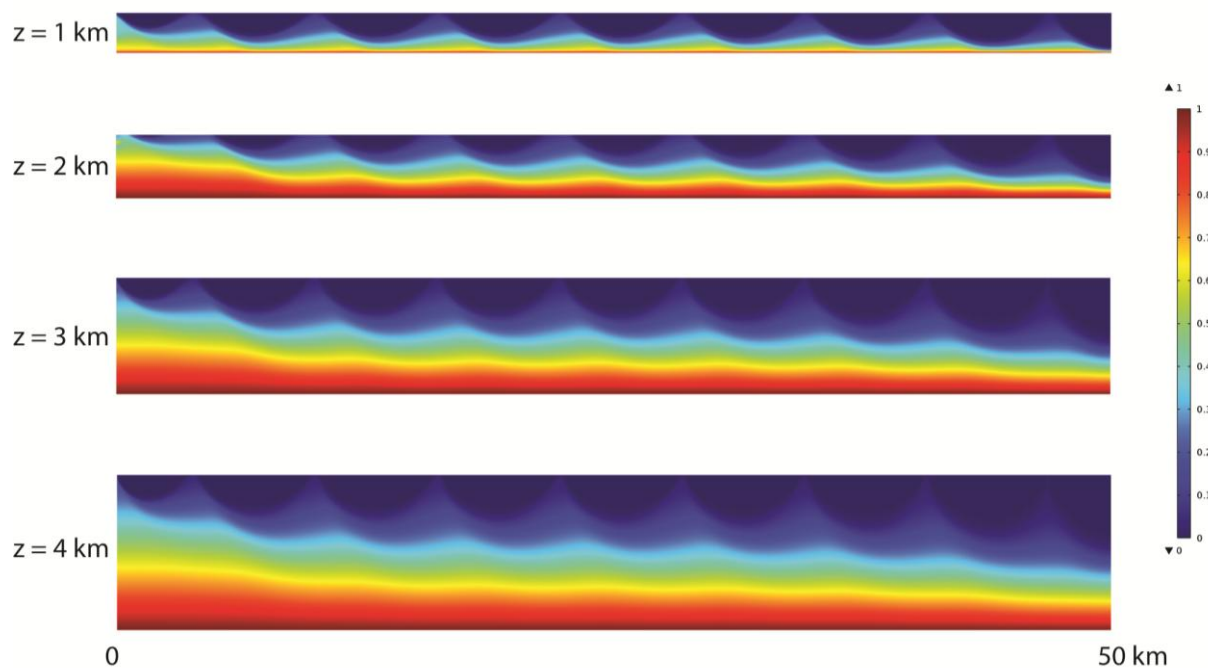


Figure 60. Distribution des concentrations sous l'influence du système d'écoulement pour 4 profondeurs différentes. L'échelle de couleur représente la concentration (normalisée par celle de l'eau de mer).

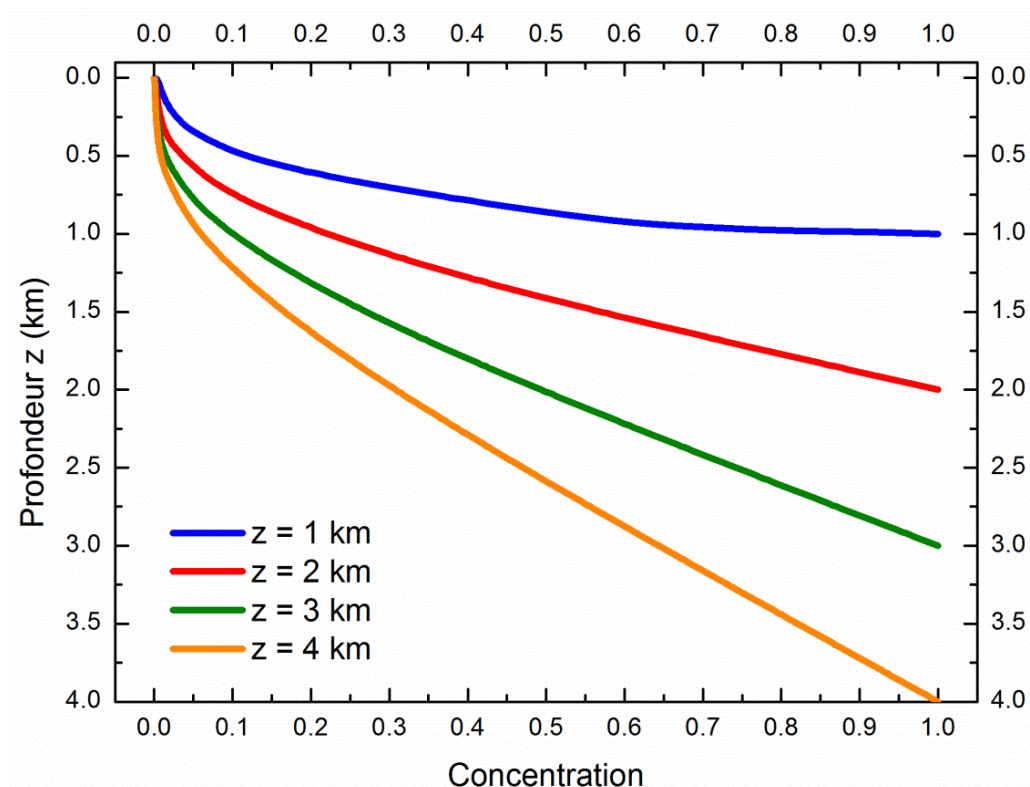


Figure 61. Profils verticaux de la concentration moyenne relative à celle de l'eau de mer au sein du système pour les 4 profondeurs différentes.

La caractérisation de ces processus de lessivage pourrait aussi être effectuée à l'aide de l'étude des flux au sein du système. La Figure 62 présente les flux au sein du système pour deux modèles différents avec des profondeurs de 1 et 4 km, où le flux correspond au produit de la concentration et de la vitesse en ce point. Ces flux semblent caractériser les zones productives en termes de lessivage. Les flux les plus importants paraissent être localisés à une certaine profondeur au sein du système d'écoulement et semblent correspondre à la zone de transition identifiée dans les profils de concentration.

Néanmoins, il convient de remarquer à ce stade que ces simulations ont été réalisées en régime permanent dans le but d'initier des perspectives de travaux pour la compréhension de l'histoire de l'empreinte marine. Or, la condition de concentration imposée à la base du modèle dans ces modélisations équivaut à considérer un stock infini, ce qui ne semble pas réaliste (par rapport à une transgression marine qui est un événement « ponctuel » sur des temps géologiques). C'est pourquoi dans un second temps il convient de simuler ces processus en condition transitoire afin d'observer l'évolution de cette signature marine dans le temps.

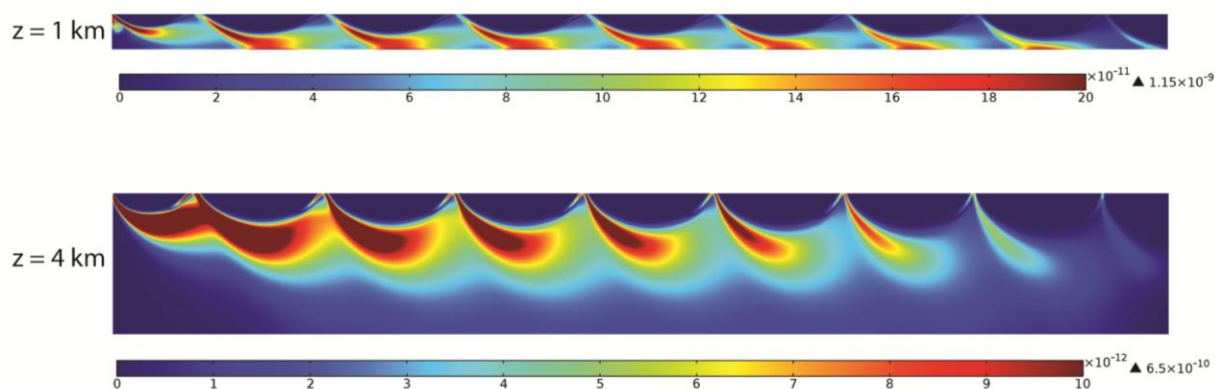


Figure 62. Flux au sein du système pour les profondeurs de 1 et 4 km.

Une première modélisation en régime transitoire a été réalisée dans cette optique, où les résultats du modèle de profondeur de 2 km (Figure 60) ont été pris comme état initial. Cette modélisation permet donc de suivre l'évolution de la signature marine (après son introduction et sa diffusion au sein du système). Les résultats de cette simulation sont synthétisés dans la Figure 63 qui montre l'évolution de la signature marine sous l'influence des écoulements sur une période de 2 Ma. La persistance de cette signature marine sur une telle échelle de temps (millions d'années) et présente

dans des concentrations non négligeables sur toute la longueur du système est en accord avec les échelles de temps de circulation de ces fluides identifiés au sein du Massif Armoricaïn (Chapitre 2). L'évolution de cette signature peut aussi être observée par le suivi de la variation du profil vertical de la concentration moyenne du système au cours du temps. La Figure 64 traduit les résultats précédents (Figure 63) sous la forme d'un profil vertical de la concentration moyenne du milieu et permet de visualiser l'évolution de la concentration moyenne du système, mais aussi d'observer la distribution au cours du temps. Sous l'effet de l'écoulement régional alimenté par les eaux météoriques, le profil de concentration évolue en fonction du temps. Du fait du processus de lessivage par les eaux météoriques, la concentration moyenne en chlorure (ou signature marine) au sein du système diminue avec le temps. Après un lessivage de l'ordre de 2 millions d'années, la concentration à la base du système a été divisée d'environ un ordre de grandeur.

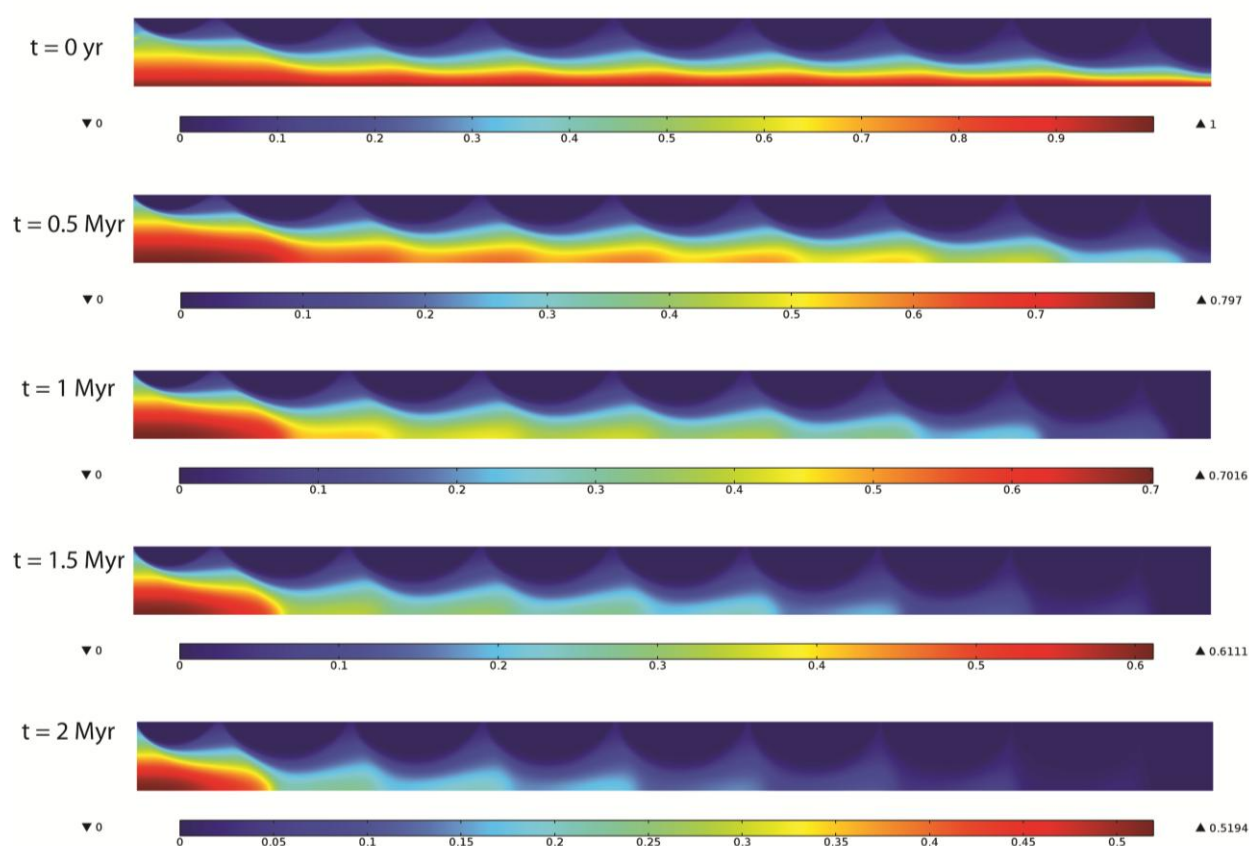


Figure 63. Évolution de la signature marine en fonction du temps sur une période de 2 Ma pour différents pas de temps (0, 0.5, 1, 1.5 et 2 Myr). L'échelle de couleur représente la concentration (normalisée par celle de l'eau de mer).

Enfin, la Figure 65 (image de droite) propose un zoom sur le profil de concentration sur les 500 premiers mètres de profondeur du système à des temps de 1 et 2Ma. Ce zoom permet d'obtenir des profils de concentration comparables au profil en termes d'ordre de grandeur de profondeur et de concentration avec les données de terrain à l'échelle du Massif Armoricaïn Figure 65 (image de gauche). En effet, l'ordre de grandeur des concentrations (autour de 50 mg/L pour une profondeur de 300m) et la forme du profil simulé semblent présenter de bonnes similitudes avec les concentrations mesurées. Cette piste intéressante mériterait donc d'être creusée dans le but de s'approcher du profil observé à l'échelle régionale des aquifères du Massif Armoricaïn en s'appuyant sur ce même type de simulation.

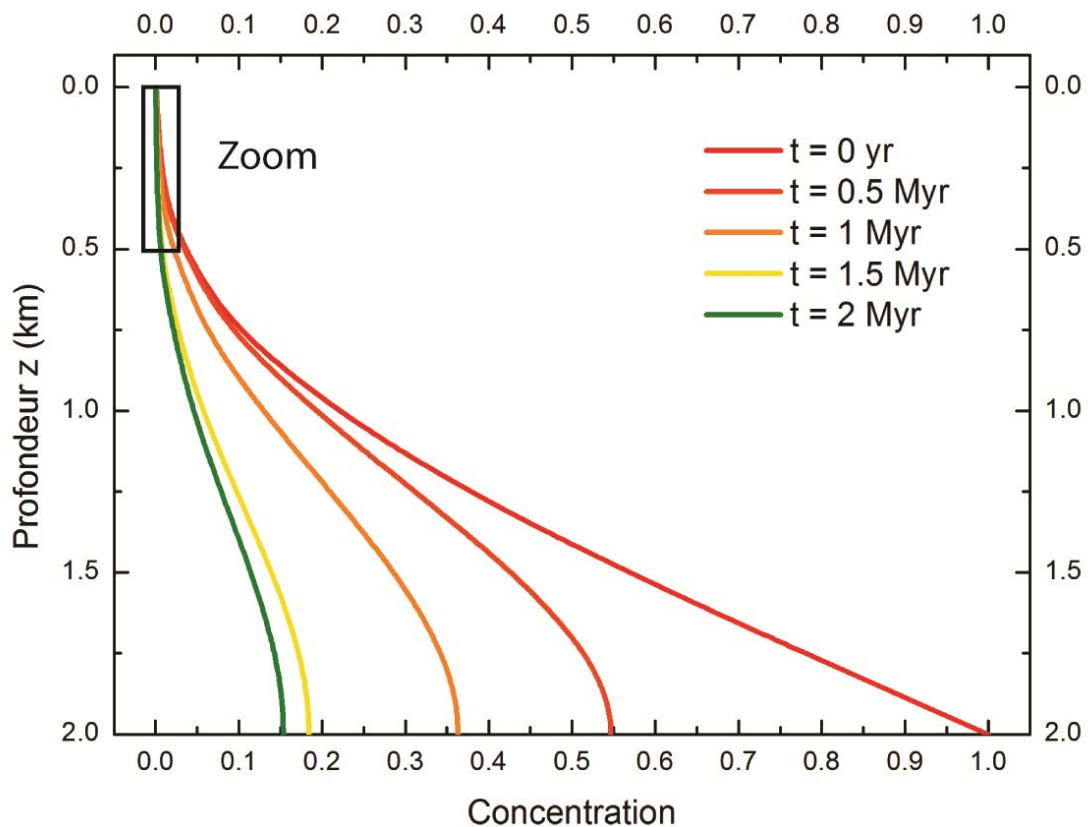


Figure 64. Profils verticaux de la concentration moyenne au sein du système de profondeur de 2 km à différents pas de temps entre 0 et 2 Ma.

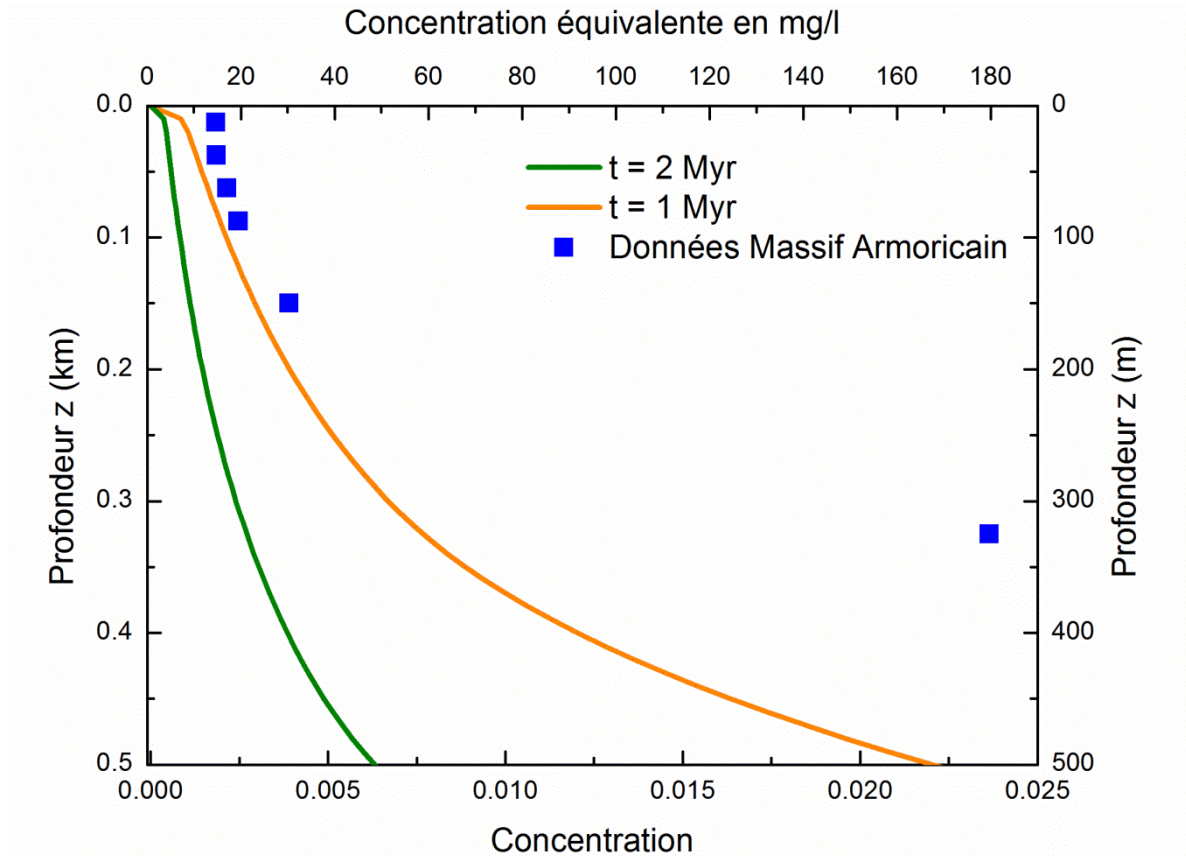


Figure 65. Carrés bleus : Profil actuel de concentration moyen en fonction de la profondeur du Massif Armoricain (données issues de la Figure 19 de la section 2.2). Courbes orange et verte : Zoom sur le profil vertical de concentration moyenne sur les 500 premiers mètres de profondeur du domaine à des temps de 1 et 2Ma.

Ces prémices de modélisation numérique à grande échelle ouvrent des perspectives pour la compréhension de l'histoire hydrogéologique des aquifères du Massif Armoricain. À partir de ces premiers résultats, il peut être envisagé de tester différents paramètres (propriétés hydrogéologiques, topographie) et différentes conditions climatiques (variations de la recharge, etc.) dans le but de tester des hypothèses, d'apporter des contraintes sur le système hydrogéologique et d'affiner l'histoire hydrogéologique tout en mettant en avant les facteurs/processus déterminants qui ont amené le système à l'état actuel.

Chapitre 5. Indices paléoclimatiques dans les eaux souterraines: Apport de la modélisation numérique à densité variable

Chapitre 6. Conclusion générale

Cette thèse a permis d'aborder la question de l'impact des variations climatiques sur les ressources hydrogéologiques à travers différentes problématiques. 1) L'impact des grands événements climatiques passés par l'analyse à l'échelle régionale de traceurs environnementaux issus de la ressource en eau souterraine actuelle 2) L'impact du climat et de ses variations sur l'ensemble du système hydrogéologique à travers l'étude des relations entre l'eau souterraine et les compartiments de surface d'une part (Chapitre 3) et l'océan (Chapitre 4).

L'analyse de la distribution à grande échelle des données géochimiques a mis en évidence l'empreinte des événements climatiques majeurs passés (à l'échelle du million d'années) au sein de la ressource en eau souterraine actuelle. L'empreinte marquée de ces événements souligne la sensibilité de la ressource hydrogéologique aux variations climatiques. L'étude et la caractérisation de ces informations paléohydrogéologiques favorisent la compréhension du fonctionnement du système hydrogéologique à grande échelle spatiale et temporelle.

L'étude des impacts climatiques sur la ressource en eau souterraine constitue donc un point essentiel dans la gestion de la ressource. Les études hydrogéologiques menées à l'aide de la modélisation numérique visent à répondre à certaines de ces questions relatives aux problématiques de gestion des ressources hydrogéologiques face aux impacts climatiques.

L'étude concernant la salinisation des aquifères découle de la mise en évidence de la persistance du signal sur plusieurs millions d'années des intrusions d'eau de mer. Dans un souci de gestion qualitative de la ressource en eau souterraine pour son exploitation, il est indispensable d'identifier et de quantifier les processus jouant un rôle dans la salinisation des aquifères afin de prévenir la dégradation de la qualité de l'eau souterraine. L'étude née de cette idée a permis de mettre en évidence l'importance de prendre en compte les systèmes de surface comme les rivières dans les processus responsables de la salinisation des aquifères du fait de leurs interactions avec l'océan. Les résultats de cette étude ouvrent certaines perspectives d'une part à l'échelle régionale du Massif Armoricaire, mais aussi à l'échelle mondiale, où il serait envisageable

de produire une cartographie actuelle des zones potentiellement impactées par les rivières sous l'influence de l'océan. Dans le cadre du changement climatique et de l'augmentation du niveau de la mer attendue, ces résultats pourraient servir de base pour étudier l'effet de l'augmentation du niveau marin sur la salinisation potentielle de nouvelles zones. Une cartographie pourrait être produite afin d'anticiper les problèmes liés à la salinisation des eaux souterraines notamment dans le cadre de son exploitation en tant qu'eau potable.

D'autre part, dans un souci de gestion quantitative de la ressource en eau souterraine, les questions relatives au changement climatique et à ses impacts sur le cycle hydrologique nécessitent d'être étudiées. Le réchauffement actuel s'accéléralant du fait des activités anthropiques, la question principale concernant le changement climatique correspond donc à son impact sur le cycle hydrologique. En effet, dans de nombreux cas, les données montrent que le cycle de l'eau est déjà impacté. Néanmoins, les travaux sur l'impact de ces changements sur la ressource en eau souterraine sont toujours limités. La première cause étant sans doute la nécessité d'avoir une base de données hydrométéorologique conséquente couvrant une longue période de temps afin de pouvoir caractériser ce changement climatique. De plus, la prédiction des effets à long terme de la dynamique du système reste difficile du fait des limitations propres aux modèles et des nombreux paramètres qui contrôlent le système climatique. Néanmoins, un modèle numérique du système hydrogéologique testé pour des scénarios possibles de changement climatique reste un outil puissant dans le but de prévenir la détérioration de la ressource en eau. L'étude du chapitre 3 met en avant l'impact direct du changement climatique et des diverses activités anthropiques sur la ressource en eau souterraine. Du fait de l'étroit lien entre l'eau souterraine et les compartiments de surface comme les zones humides, l'impact se généralise à l'ensemble du système hydrogéologique. Sur le long terme pour le système étudié, l'impact des activités anthropiques indirectes (changement climatique et son réchauffement) semble prédominant en comparaison avec les activités anthropiques directes (pompages, drainage...). Il devient donc indispensable de cerner les futurs problèmes en eau et environnementaux auxquels nous serons soumis à travers des études focalisées en utilisant des bases de données météorologiques et hydrologiques de bonne qualité. En effet, la préservation d'écosystèmes qui comportent des ressources en eau critiques passe par la protection de zones telles que les zones de recharge, les zones humides, etc.

d'où une nécessité de contribuer à l'amélioration de la gestion des eaux souterraines. Malgré les incertitudes la constitution de nouvelles stratégies de gestion de la ressource en eau basée sur la modélisation peut permettre d'anticiper, de prévenir et/ou de minimiser les causes du changement climatique et d'atténuer ses effets négatifs. Enfin, si les conséquences sur le système hydrogéologique résultent à la fois du changement climatique et des évolutions socio-économiques, la pérennité de la ressource dépend de la mise en commun des compétences de chercheurs des divers domaines avec celles des décisionnaires.

Ces divers résultats soulignent l'importance de la sensibilité des ressources hydrogéologiques aux variations climatiques, sur différentes échelles de temps et à travers différents mécanismes. Ces travaux ouvrent de larges perspectives, allant de la capacité à contraindre le fonctionnement hydrogéologique des aquifères de socle à grande échelle spatiale et temporelle, à la prévision de l'impact des activités anthropiques directes et indirectes sur la ressource en souterraine et ses compartiments associés, à la quantification et à la cartographie des zones (aquifères) potentiellement influencées à l'heure actuelle et qui pourront l'être par la salinité du système fluvial, en passant par l'étude ou la collecte de nouvelles données géochimiques à l'échelle régionale et de la modélisation numérique des écoulements (en 3D, en 2D, à densité variable...).

Conclusion générale

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ANNEXE 2 (Modèle dernière page de thèse)

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